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(54) Title of the Invention: IMAGE RECORDING BODY AND METHOD
FOR MANUFACTURING THE SAME

# (57) Abstract:

PROBLEM TO BE SOLVED: To provide an image recording body which is excellent in a property of printing letters and is not only capable of enhancing security against forgery, an alteration or the like but also capable of preventing scratches or wrinkles from being generated more compared with a conventional method and, further, capable of preventing blocking from being formed during a storage in an extended period of time and, still further, not only capable of preventing the forgery or the alteration without impairing the property of printing the letters and durability but also capable of being readily prepared in a card form.

SOLUTION: In the image recording body containing at least one information carrying layer 2 which carries one item selected from at least identification information and bibliographic information on a substrate 1, a reflection layer 5 and/or a bead-containing layer 3 which contains beads and an actinic ray-cured layer 4 are provided on the information—carrying layer 2 in the stated order, and a printed layer 7, a reflection layer 5 and/or a bead-containing layer 3 which contains beads and an actinic ray-cured layer 4 are provided on the information—carrying layer 2 in the stated order.

# [Claims]

# [Claim 1]

An image recording body containing at least one information-carrying layer which carries one item selected from at least identification information and bibliographic information on a substrate, being characterized in that a reflection layer and/or a bead-containing layer which contains beads and an actinic ray-cured layer are provided on the information-carrying layer in the stated order.

# [Claim 2]

An image recording body containing at least one information-carrying layer which carries one item selected from at least identification information and bibliographic information on a substrate, being characterized in that a printed layer, a reflection layer and/or a bead-containing layer which contains beads and an actinic ray-cured layer are provided on the information-carrying layer in the stated order. [Claim 3]

The image recording body according to Claim 1 or 2, being characterized in that the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%.

# [Claim 4]

The image recording body according to Claim 1 or 2, being characterized in that the actinic ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5.

# [Claim 5]

The image recording body according to any one of Claims 1 to 4, being characterized in that a reflection layer is provided on the side of the substrate of the bead-containing layer; and an adhesion force between the bead-containing layer of the beads between layer reflection or the and bead-containing layer and the reflection layer is smaller than any other adhesion forces between any two of other layers. [Claim 6]

The image recording body according to any one of Claims 1 to 4, being characterized in that an adhesion force between the bead-containing layer and a layer adjacent to the bead-containing layer is larger than that of beads in the bead-containing layer.

# [Claim 7]

The image recording body according to any one of Claims 1 to 6, being characterized in that the reflection layer contains a pearl pigment.

#### [Claim 8]

The image recording body according to any one of Claims 1 to 6, being characterized in that a colorless or colored uneven picture pattern is provided in the printed layer.

[Claim 9]

The image recording body according to Claim 8, being characterized in that the uneven picture pattern contains a

pearl pigment different from that according to Claim 7 in terms of interference.

# [Claim 10]

The image recording body according to any one of Claims 1 to 9, being characterized in that at least one type of reflection layer is provided on the side of the substrate of the bead-containing layer; the bead-containing layer contains beads; and the reflection layer appears to be in a character or logo shape.

#### [Claim 11]

The image recording body according to Claim 10, being characterized in that at least one portion of the reflection layer which appears to be in a character or logo shape is overlapped with portions of the identification information and the bibliographic information.

#### [Claim 12]

The image recording body according to any one of Claims 1 to 9, being characterized in that at least one type of reflection layer is provided on the side of the substrate of the bead-containing layer; and a frequency of a quantity of existence of beads in the bead-containing layer appears to be in a character or logo shape.

#### [Claim 13]

The image recording body according to Claim 12, being characterized in that at least one portion of the

bead-containing layer which appears to be in the character or logo shape is overlapped with the portions of the identification information and the bibliographic information.

[Claim 14]

The image recording body according to any one of Claims

1 to 13, being characterized in that the beads in the

bead-containing layer do not exist in a peripheral portion

thereof in a card shape.

#### [Claim 15]

The image recording body according to any one of Claims 1 to 13, being characterized in that at least one portion of the identification information and bibliographic information is identified by a recognizing apparatus; and the bead-containing layer or beads do not exist in the portion recognized by the recognizing apparatus.

#### [Claim 16]

A method for producing an image recording body, being characterized in that at least an actinic ray-cured layer is provided on the side having releasability of a releasable support; and, after forming the actinic ray-cured layer, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend. [Claim 17]

A method for producing an image recording body, being

characterized in that at least an actinic ray-cured layer is provided on the side having releasability of a releasable support; and, after forming the actinic ray-cured layer, a transfer foil in which an interlayer and an adhesive layer are successively provided is transferred onto an adherend in which a protective layer, a bead-containing layer containing beads and/or a reflection layer have previously been transferred onto an information-carrying layer.

# [Claim 18]

A method for producing an image recording body, being characterized in that, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray; and, then, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend.

# [Claim 19]

A method for producing an image recording body, being characterized in that, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray; and, then, a transfer foil in which an adhesive layer, a

bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend.

[Claim 20]

The method for producing the image recording body according to any one of Claims 16 to 19, being characterized in that the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%.

[Claim 21]

The method for producing the image recording body according to any one of Claims 16 to 19, being characterized in that the actinic ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5.

[Detailed Description of the Invention]

[0001]

[Technical Field to which the Invention Belongs]

The present invention relates to an image recording body which is suitably applied to a contact or non-contact type electronic or magnetic card, sheet or the like which stores personal information and the like for which security is demanded to prevent forgery, an alteration or the like, and also to a method for producing the image recording body.

[Prior Art]

[0002]

In recent years, contact or non-contact-type electronic

or magnetic cards and the like have increasingly been issued in the field of service industry such as banks, business organizations, educational institutions, public agencies, and the like. Personal information is recorded on cash cards, employee cards, company membership cards, membership cards, student cards, aliens registration cards, various types of license cards and the like which are employed in various fields. Accordingly, these cards are subjected to a security treatment so that forgery as well as an alteration is not readily carried out.

[0003]

For example, a hologram layer is provided onto a card substrate and a special image is formed so that a portrait, a name, an issue date and the like are not readily imitated. The image is generally called as a diffraction grating image or a hologram image. The hologram image is formed employing interference infringes obtained by overlapping waves which are reflected from a body or transmitted through a body with the reference waves. By employing the hologram image, some image of the body is stereoscopically reproduced. A surface of the hologram layer is covered with a protective sheet having high light transmission. Employed as the protective sheet are transparent resins and the like which are employed for lamination, and the hologram layer which exhibits less strength than the card substrate is covered with the protective sheet

and thereby protected.

[0004]

[0005]

[Problems that the Invention is to Solve]

As described above, the hologram layer is provided on the card substrate. On this occasion, there is a fear in that the hologram layer and a portion on which information is recorded are peeled off and, then, the forgery or the alteration is performed by providing another information, or information and the hologram layer are simultaneously peeled off and, then, the forgery or the alteration is performed by providing both another information and the hologram layer.

Further, according to a method for producing a card using a conventional system, for instance, a laminate film and a hologram sheet are separately formed and, after passing through a laminating process, an extra laminate film is trimmed. Specifically for a photo-bearing company membership card, a driver's license card or the like, in order to ensure positive identification with a flawless image, it is necessary that, during a production process, formation of a scratch as well as a wrinkling on a surface of the card is minimized and it is further necessary to take heed of preventing the hologram sheet wound in a roll from forming blocking during a storage in an extended period of time.

[0006]

Then, invention the solved present the has above-described problems and has an object to provide an image recording body which is excellent in a property of printing letters and is not only capable of enhancing security against an alteration or the like but also capable of forgery, preventing scratches or wrinkles from being generated more compared with a conventional method and, further, capable of preventing blocking from being formed during a storage in an extended period of time and, still further, not only capable of preventing the forgery or the alteration without impairing the property of printing the letters and durability but also capable of being readily prepared in a card form, and a method for producing the image recording body.

[0007]

[Means for Solving the Problems]

In order to solve the aforementioned problems and to achieve an object, the present invention is constituted as described below.

[8000]

The invention as described in Claim 1 is "an image recording body having at least one information-carrying layer which carries one item selected from at least identification information and bibliographic information on a substrate, being characterized in that a reflection layer and/or a bead-containing layer which contains beads and an actinic

ray-cured layer are provided on the information-carrying layer in the stated order".

[0009]

[0011]

According to the invention as described in Claim 1, by providing the reflection layer and/or the bead-containing layer which contains beads and the actinic ray-cured layer on the information-carrying layer in the stated order, a utilization efficiency of light can be enhanced without impairing printability, to thereby enhance security against the forgery, the alteration or the like.

The invention as described in Claim 2 is "an image recording body having at least one information-carrying layer which carries one item selected from at least identification information and bibliographic information on a substrate, being characterized in that a printed layer, a reflection layer and/or a bead-containing layer which contains beads and an actinic ray-cured layer are provided on the information-carrying layer in the stated order".

According to the invention as described in Claim 2, by providing the printed layer, the reflection layer and/or the bead-containing layer which contains beads and the actinic ray-cured layer on the information-carrying layer in the stated order, a utilization efficiency of light can be enhanced

without impairing printability, to thereby enhance security against the forgery, the alteration or the like. [0012]

The invention as described in Claim 3 is "the image recording body according to Claim 1 or 2, being characterized in that the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%".

According to the invention as described in Claim 3, the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%" and generation of scratches or wrinkles can be prevented compared with a conventional method and, further, blocking to be formed during a storage in an extended period of time can be prevented.

[0014]

The invention as described in Claim 4 is "the image recording body according to Claim 1 or 2, being characterized in that the actinic ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5".
[0015]

According to the invention as described in Claim 4, the actinic ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5 and generation of scratches or wrinkles can be prevented compared with a conventional method and, further, blocking to be formed during

a storage in an extended period of time can be prevented. [0016]

The invention as described in Claim 5 is "the image recording body according to any one of Claims 1 to 4, being characterized in that a reflection layer is provided on the side of the substrate of the bead-containing layer; and an adhesion force between the bead-containing layer and the reflection layer or between beads of the bead-containing layer and the reflection layer is smaller than any other adhesion forces between any two of other layers".

According to the invention as described in Claim 4, even when the forgery or the alteration is tried to be performed by peeling the information-carrying layer, the bead-containing layer and the reflection layer are separated from each other, or beads in the bead-containing layer and the reflection layer are separated from each other and, then, the bead-containing layer is broken or beads therein are dropped off such that it can not be used again and, therefore, the forgery or the alteration can not be performed.

The invention as described in Claim 6 is "the image recording body according to any one of Claims 1 to 4, being characterized in that an adhesion force between the bead-containing layer and layer a adjacent the to

[0018]

bead-containing layer is larger than that of beads in the bead-containing layer".

[0019]

According to the invention as described in Claim 6, even when the forgery or the alteration is tried to be performed by peeling the information-carrying layer, beads in the bead-containing layer are dropped off therefrom such that it is broken and can not be used again and, therefore, the forgery or the alteration can not be performed.

[0020]

The invention as described in Claim 7 is "the image recording body according to any one of Claims 1 to 6, being characterized in that the reflection layer contains a pearl pigment".

[0021]

According to the invention as described in Claim 7, by allowing the reflection layer to contain the pearl pigment, the security against the forgery, the alteration or the like can be enhanced.

[0022]

The invention as described in Claim 8 is "the image recording body according to any one of Claims 1 to 6, being characterized in that a colorless or colored uneven picture pattern is provided in the printed layer".

[0023]

According to the invention as described in Claim 8, by providing the colorless or colored uneven picture pattern in the printed layer, the security against the forgery, the alteration or the like can be enhanced.

[0024]

The invention as described in Claim 9 is "the image recording body according to Claim 8, being characterized in that the uneven picture pattern contains a pearl pigment different from that according to Claim 7 in terms of interference".

[0025]

According to the invention as described in Claim 9, by allowing the uneven picture pattern to contain a pearl pigment different from that according to Claim 7 in terms of interference, the security against the forgery, the alteration or the like can be enhanced.

[0026]

The invention as described in Claim 10 is "the image recording body according to any one of Claims 1 to 9, being characterized in that at least one type of reflection layer is provided on the side of the substrate of the bead-containing layer; the bead-containing layer contains beads; and the reflection layer appears to be in a character or logo shape".

[0027]

According to the invention as described in Claim 10, by

allowing the bead-containing layer to contain beads and by allowing a character or logo shape to be formed in the reflection layer, a security treatment can be performed such that the forgery or the alteration can easily and surely be prevented.

[0028]

The invention as described in Claim 11 is "the image recording body according to Claim 10, being characterized in that at least one portion of the reflection layer which appears to be in a character or logo shape is overlapped with portions of the identification information and the bibliographic information".

[0029]

According to the invention as described in Claim 11, by allowing at least one portion of the reflection layer which appears to be in the character or logo shape to be overlapped with portions of the identification information and the bibliographic information, the forgery or the alteration can even more surely be prevented.

[0030]

The invention as described in Claim 12 is "the image recording body according to any one of Claims 1 to 9, being characterized in that at least one type of reflection layer is provided on the side of the substrate of the bead-containing layer; and a frequency of a quantity of existence of beads in

the bead-containing layer appears to be in a character or logo shape".

[0031]

According to the invention as described in Claim 13, the frequency of the quantity of existence of the beads in the bead-containing layer appears to be in the character or logo shape and, then, a security treatment can be performed such that the forgery or the alteration can easily and surely be prevented.

[0032]

The invention as described in Claim 13 is "the image recording body according to Claim 12, being characterized in that at least one portion of the bead-containing layer which appears to be in the character or logo shape is overlapped with the portions of the identification information and the bibliographic information".

[0033]

According to the invention as described in Claim 13, by allowing at least one portion of the bead-containing layer which appears to be in the character or logo shape to be overlapped with the portions of the identification information and the bibliographic information, the forgery or the alteration can even more surely be prevented.

[0034]

The invention as described in Claim 14 is "the image

recording body according to any one of Claims 1 to 13, being characterized in that the beads in the bead-containing layer do not exist in a peripheral portion thereof in a card shape".

[0035]

According to the invention as described in Claim 14, since the beads in the bead-containing layer do not exist at a position at which a punching press performs punching, at the time of performing a punching-out operation in a card shape by the punching press, the beads are not dropped off or durability of the punching press is not impaired.

[0036]

The invention as described in Claim 15 is "the image recording body according to any one of Claims 1 to 13, being characterized in that at least one portion of the identification information and bibliographic information is identified by a recognizing apparatus; and the bead-containing layer or beads do not exist in the portion recognized by the recognizing apparatus".

[0037]

According to the invention as described in Claim 15, since the bead-containing layer or the beads do not exist in the portion recognized by the recognizing apparatus, it is possible to read at least one portion of the identification information and the bibliographic information and, then, to easily inspect whether the forgery or the alteration has been

made or not from the thus-read information. [0038]

The invention as described in Claim 16 is "a method for producing an image recording body, being characterized in that at least an actinic ray-cured layer is provided on the side having releasability of a releasable support; and, after forming the actinic ray-cured layer, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend".

According to the invention as described in Claim 16, after forming the actinic ray-cured layer, by transferring the transfer foil in which the bead-containing layer containing beads and/or the reflection layer, the interlayer and the adhesive layer are successively provided onto an adherend, a utilization efficiency of light can be enhanced without impairing printability, to thereby enhance security against

[0040]

forgery, alteration or the like.

The invention as described in Claim 17 is "a method for producing an image recording body, being characterized in that at least an actinic ray-cured layer is provided on the side having releasability of a releasable support; and, after forming the actinic ray-cured layer, a transfer foil in which

an interlayer and an adhesive layer are successively provided is transferred onto an adherend in which a protective layer, a bead-containing layer containing beads and/or a reflection layer have previously been transferred onto an information-carrying layer".

[0041]

According to the invention as described in Claim 18, after forming the actinic ray-cured layer, by transferring the transfer foil in which the interlayer and the adhesive layer are successively provided onto the adherend in which the protective layer, the bead-containing layer containing beads and/or the reflection layer have previously been transferred onto the information-carrying layer, a utilization efficiency of light can be enhanced without impairing printability, to thereby enhance security against forgery, alteration or the like.

[0042]

The invention as described in Claim 18 is "a method for producing an image recording body, being characterized in that, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray; and, then, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are

successively provided is transferred onto an adherend". [0043]

According to the invention as described in Claim 18, after the actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray, by transferring the transfer foil in which the bead-containing layer containing beads and/or the reflection layer, the interlayer and the adhesive layer are successively provided onto the adherend, a utilization efficiency of light can be enhanced without impairing printability, to thereby enhance security against forgery, alteration or the like.

The invention as described in Claim 19 is "a method for producing an image recording body, being characterized in that, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray; and, then, a transfer foil in which an adhesive layer, a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend".

[0045]

According to the invention as described in Claim 19, an actinic ray-cured layer is formed by exposing the thus-applied

actinic ray-curable liquid to actinic ray and, then, by transferring the transfer foil in which the adhesive layer, the bead-containing layer containing beads and/or the reflection layer, the interlayer and the adhesive layer are successively provided onto the adherend, a utilization efficiency of light can be enhanced without impairing printability, to thereby enhance security against forgery, alteration or the like.

### [0046]

The invention as described in Claim 20 is "the method for producing the image recording body according to any one of Claims 16 to 19, being characterized in that the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%".

#### [0047]

According to the invention as described in Claim 20, the actinic ray-cured layer has a breaking elongation in the range of from 5 to 90%, generation of scratches or wrinkles can more be prevented compared with a conventional method and, further, blocking to be formed during a storage in an extended period of time can be prevented.

# [0048]

The invention as described in Claim 21 is "the method for producing the image recording body according to any one of Claims 16 to 19, being characterized in that the actinic

ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5".

According to the invention as described in Claim 21, the actinic ray-cured layer has a static surface friction coefficient in the range of from 0.01 to 0.5, generation of scratches or wrinkles can more be prevented compared with a conventional method and, further, blocking to be formed during a storage in an extended period of time can be prevented. [0050]

[Mode for Carrying Out the Invention]

Hereinafter, embodiments of an image recording body and a method for producing the image recording body according to the present invention will be described with reference to drawings; however, the present invention is not limited to these embodiments, description and drawings.

[0051]

FIGS. 1 to 7 are each a diagram showing a layer constitution of an image recording body. The image recording body of the embodiment as shown in FIG. 1 has an information-carrying layer 2, a bead-containing layer containing beads 3 and an actinic ray-cured layer 4 in the state order on a substrate 1. In the information-carrying layer 2, at least one item selected from identification information such as profile image and the like and bibliographic information

such as specified number, signal and the like is carried. By allowing the bead-containing layer 3 to be provided on the information-carrying layer 2, a utilization efficiency of light can be enhanced by retroreflection of the bead-containing layer 3 without impairing printability.

[0052]

Further, since the actinic ray-cured layer 4 has a breaking elongation in the range of from 5 to 90%, a utilization efficiency of light can be enhanced without impairing printability and not only security against the forgery, the alteration or the like is enhanced but also generation of scratches or wrinkles is more prevented compared with a conventional method and, further, blocking to be formed during a storage in an extended period of time can be prevented. [0053]

Further, the actinic ray-cured layer 4 has a static surface friction coefficient in the range of from 0.01 to 0.5, a utilization efficiency of light can be enhanced without impairing printability and not only security against the forgery, the alteration or the like is enhanced but also generation of scratches or wrinkles is more prevented compared with a conventional method and, further, blocking to be formed during a storage in an extended period of time can be prevented. [0054]

The image recording body of the embodiment as shown in

FIG. 2 is constituted in a same manner as in the embodiment as shown in FIG. 1 except for having a reflection layer 5 in place of the bead-containing layer 3. The reflection layer 5 is an optically variable device layer of hologram or the like and preferably uses a pearl pigment of an interference type. [0055]

The image recording body of the embodiment as shown in FIG. 3 is constituted in a same manner as in the embodiment as shown in FIG. 1 and has a reflection layer 5 on the side of the substrate of the bead-containing layer 3. By providing not only the reflection layer 5 which is an optically variable device layer of hologram or the like but also the bead-containing layer 3, a utilization efficiency of light can further be enhanced by the retroreflection of the bead-containing layer 3.

[0056]

An adhesion force between the bead-containing layer 3 and the reflection layer 5 or that between beads in the bead-containing layer 3 and the reflection layer 5 is set to be smaller than that between any two of other layers and, even when the forgery or the alteration is attempted by peeling the information-carrying layer 2 off, the bead-containing layer 3 and the reflection layer 5 are separated from each other, or the beads in the bead-containing layer 3 and the reflection layer 5 are separated from each other, the

bead-containing layer 3 is broken or the beads therein drop off such that the bead-containing layer 3 can not be used again and, therefore, the forgery or the alteration can not be performed.

[0057]

Further, an adhesion force between the bead-containing layer 3 and a layer adjacent to the bead-containing layer 3 is set to be larger than that of beads in the bead-containing layer 3 and, even when the forgery or the alteration is tried to be performed by peeling the information-carrying layer 2, the beads are separated from the bead-containing layer 3 and, then, the bead-containing layer 3 is broken such that the bead-containing layer 3 can not be used again and, therefore, the forgery or the alteration can not be performed.

The image recording body of the embodiment as shown in FIG. 4 is constituted in a same manner as in the embodiment as shown in FIG. 1 and has a printed layer 7 between the information-carrying layer 2 and the bead-containing layer 3. The image recording body of the embodiment as shown in FIG. 5 is constituted in a same manner as in the embodiment as shown in FIG. 2 and has the printed layer 7 between the information-carrying layer 2 and the reflection layer 5. The image recording body of the embodiment as shown in FIG. 6 is constituted in a same manner as in the embodiment as shown in

FIG. 3 and has the printed layer 7 between the information-carrying layer 2 and the reflection layer 5. [0059]

It is preferable to use a pearl pigment of an interference type in the reflection layer 5. The printed layer 7 has an uneven colorless or colored picture pattern. The pearl pigment to be used in the printed layer 7 may be different from that to be used in the reflection layer 5. By means of the reflection layer 5 and the printed layer 7, the forgery or the alteration can more surely be prevented.

The image recording body as shown in each of FIGS. 7 to 9 is constituted in a same manner as in the embodiment as shown in FIG. 3. In the embodiment as shown in FIG. 7, the reflection layer 5 is provided in correspondence with a portion of the bead-containing layer 3 while, in the embodiment as shown in FIG. 8, the bead-containing layer 3 is provided in correspondence with a portion of the reflection layer 5 and, further, in the embodiment as shown in FIG. 9, the reflection layer 5 and the bead-containing layer 3 are provided in accordance with each other and, by these arrangements, a character or logo shape is formed and, then, a security treatment can be performed such that the forgery or the alteration can easily and surely be prevented.

[0061]

As described above, by having at least one type of reflection layer 5 on the side of the substrate of the bead-containing layer 3, allowing the bead-containing layer 3 to contain beads and allowing the reflection layer 5 to appear to be in a character or logo shape, a security treatment can be performed such that the forgery or the alteration can easily and surely be prevented. Further, by allowing at least one portion of the reflection layer 5 which appears to be in the character or logo shape to be overlapped with portions of the identification information and the bibliographic information, the forgery or the alteration can more surely be prevented. [0062]

Further, by having at least one type of reflection layer 5 on the side of the substrate of the bead-containing layer 3, allowing frequency of a quantity of existence of the beads in the bead-containing layer 3 to appear to be in a character or logo shape, a security treatment can be performed such that the forgery or the alteration can easily and surely be prevented. Still further, by allowing at least one portion of the bead-containing layer 3 which appears to be in the character or logo shape to be overlapped with portions of the identification information and the bibliographic information, the forgery or the alteration can more surely be prevented. [0063]

FIGS. 10 to 13 each show a layout of the image recording

body. The embodiment as shown in FIG. 10 has identification information 30 such as facial portrait, bibliographic information 31 such as specific number or the like, an optical recognition portion 32 and, information further, bead-containing layer 3. The bead-containing layer 3 is provided such that it overlaps with a portion of the identification information 30 and a portion the bibliographic information 31 and, even when the identification information 30 and the bibliographic information 31 are tried to be falsified, since the bead-containing layer 3 is broken, the forgery or the alteration can be prevented. Still further, by not providing the bead-containing layer 3 in the optical information recognition portion 32, information recorded in the optical information recognition portion 32 is read by an information recognition apparatus, to thereby perform inspection or the like on production of the image recording body, or the forgery or the alteration.

[0064]

FIG. 11 is constituted in a same manner as in the embodiment as shown in FIG. 10, but, on the basis of a position information provided in the printed layer 7, a periphery is punched out by a punching press, to thereby form a card shape. [0065]

FIG. 12 is provided with the printed layer 7 in a same manner as in the embodiment as shown in FIG. 11 and, on the

basis of a position information of the printed layer 7, the bead-containing layer 3 is provided inside, but not in the optical information recognition portion 32 and, then, information recorded in the optical information recognition portion 32 can be read by the information recognition apparatus.

[0066]

FIG. 13 is provided with the printed layer 7 in a same manner as in the embodiment as shown in FIG. 11 and, on the basis of a position information of the printed layer 7, the reflection layer 5 is provided inside and, then, information recorded in the optical information recognition portion 32 can be read by the information recognition apparatus.

The bead-containing layer 2 in this embodiment has a reflection substrate and transparent beads aligned on the substrate and, in this embodiment, may be provided such that articles which are each previously formed in a sheet shape are laminated one on top of another at a specified position or reflection substrates are laminated one on top of another at a specified position and, then, beads are provided on the resultant laminate.

[0068]

Next, a method for producing an image recording body according to the present invention will be described. In a

first embodiment, an actinic ray-cured layer is provided on the side having releasability of a releasable support and, after forming the actinic ray-cured layer, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer, an adhesive layer are successively provided is transferred onto an adherend.

Further, in a second embodiment, an actinic ray-cured layer is provided on the side having releasability of a releasable support and, after forming the actinic ray-cured layer, a transfer foil in which an interlayer and an adhesive layer are successively provided is transferred onto an adherend in which a protective layer, a bead-containing layer containing beads and/or a reflection layer have previously been transferred onto an information-carrying layer.

Still further, in a third embodiment, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray and, then, a transfer foil in which a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend.

[0071]

Even still further, in a fourth embodiment, after applying an actinic ray-curable liquid on the side having releasability of a releasable support, an actinic ray-cured layer is formed by exposing the thus-applied actinic ray-curable liquid to actinic ray and, then, a transfer foil in which an adhesive layer, a bead-containing layer containing beads and/or a reflection layer, an interlayer and an adhesive layer are successively provided is transferred onto an adherend.

[0072]

The term "transfer foil" as used herein refers to a transparent protective foil for protecting an image; the term "transparent protective layer" as used herein refers to a transparent protective layer for protecting an image; the term "resin layer" as used herein refers to a layer containing a resin in which a breaking elongation and a friction coefficient are defined; the term "actinic curing ray resin layer" as used herein refers to a portion of types of resin layers and, although it is a preferred embodiment in the present invention, it is sometimes limited to the resin layer on a production standpoint; the term "curable protective layer-containing optically variable device layer" as used herein refers to a layer in which a cured layer and an optically variable device layer are combined with each other to be a unity; and the term "protection-imparted transfer foil" as used herein refers to

a transfer foil containing at least one layer of a resin layer (preferably of actinic ray-curable resin).
[0073]

Next, embodiments of a transparent protective transfer foil 64 are shown in FIG. 14. The transparent protective transfer foil 64 as shown in FIG. 14(a) is constituted with a transparent protective transfer layer 640 and a support 64b; the transparent protective transfer layer 640 is constituted with a peeling layer 64a1, a transparent protective layer 64a2 and an adhesive layer 64a3; the peeling layer 64a1 and the adhesive layer 64a3 are provided on respective sides of the transparent protective layer 64a2; and the peeling layer 64a1 is adhered to the support 64b. Although the transparent protective transfer foil 64 as shown in FIG. constituted in a same manner as in the transfer foil as shown in FIG. 14(a), an interlayer 64a4 is provided between the transparent protective layer 64a2 and the adhesive layer 64a3. Although the transparent protective transfer foil 64 as shown in FIG. 14(c) is constituted in a same manner as in the transfer foil as shown in FIG. 14(b), two layers of the transparent protective layers 64a2 are provided. Although the transparent protective transfer foil 64 as shown in FIG. 14(d) is constituted in a same manner as in the transfer foil as shown in FIG. 14(b), a barrier layer 64a5 is provided between the transparent protective layer 64a2 and the interlayer 64a4.

[0074]

These embodiments of the transparent protective transfer foil 64 are each transferred by allowing the transparent protective transfer layer 640 to be peeled off from a support 43b.

[0075]

Embodiments of a bead-containing transfer foil 43 are shown in FIG. 13. The bead-containing transfer foil 43 as shown in FIG. 15(a) is constituted with a bead-containing transfer layer 430 and the support 43b; the bead-containing transfer layer 430 is constituted with a peeling layer 43al, a bead-containing layer 43a2 and an adhesive layer 43a3; the peeling layer 43a1 and the adhesive layer 43a3 are provided on respective sides of the bead-containing layer 43a2; and the peeling layer 43al is adhered to the support 43b. Although the bead-containing transfer foil 43 as shown in FIG. 15(b) is constituted in a same manner as in the transfer foil as shown in FIG. 15(a), an interlayer 43a4 is provided between the adhesive layer 43a3 and the bead-containing layer 43a2. Although the bead-containing transfer foil 43 as shown in FIG. 15(c) is constituted in a same manner as in the transfer foil as shown in FIG. 15(b), a barrier layer 43a5 is provided between the bead-containing layer 43a2 and the interlayer 43a4. Although the transfer foil as shown in FIG. 15(d) is constituted in a same manner as in the transfer foil as shown in FIG. 15(c),

a transparent protective layer 43a6 is provided between the peeling layer 43a1 and the bead-containing layer 43a2. [0076]

These embodiments of the bead-containing transfer foil 43 are each transferred by allowing the bead-containing transfer layer 430 to be peeled off from the support 43b. [0077]

Next, embodiments of a resin transfer foil 66 are shown in FIG. 16. The resin transfer foil 66 as shown in FIG. 16(a) is constituted with a resin transfer layer 660 and a support 66b; the resin transfer layer 660 is constituted with a peeling layer 66al, a transparent protective layer 66a2 and an adhesive layer 66a3; the peeling layer 66al and the adhesive layer 66a3 are provided on respective sides of the transparent protective layer 66a2; and the peeling layer 66a1 is adhered to the support Although the resin transfer foil 66 as shown in FIG. 16(b) 66b. is constituted in a same manner as in the transfer foil as shown in FIG. 16(a), an interlayer 66a4 is provided between the transparent protective layer 66a2 and the adhesive layer 66a3. Although the resin transfer foil 66 as shown in FIG. 16(c) is constituted in a same manner as in the transfer foil as shown in FIG. 16(b), two layers of the transparent protective layers 66a2 are provided. Although the resin transfer foil 66 as shown in FIG. 16(d) is constituted in a same manner as in the transfer foil as shown in FIG. 16(b), a barrier layer 66a5 is provided

between the transparent protective layer 63a2 and the interlayer 63a4.

[0078]

These embodiments of the resin transfer foil 66 are each transferred by allowing the resin transfer layer 660 to be peeled off from the support 43b.

[0079]

Next, embodiments of curing-type a resin layer-containing bead-containing transfer foil 44 are shown in FIG. 17. curing-type The resin layer-containing bead-containing transfer foil 44 as shown in FIG. 17(a) is constituted with a curing-type resin layer-containing bead-containing transfer layer 440 and a support 44b; the curing-type resin layer-containing bead-containing transfer layer 440 is constituted with a peeling layer 44a1, a bead-containing layer 44a2 and an adhesive layer 44a3; the peeling layer 44a1 and the adhesive layer 44a3 are provided on respective sides of the bead-containing layer 44a2; and the peeling layer 44al is adhered to the support 44b. Although the curing-type resin layer-containing bead-containing transfer foil 44 as shown in FIG. 17(b) is constituted in a same manner as in the transfer foil as shown in FIG. 17(a), a resin layer 44a9 is provided between the peeling layer 44a1 and the bead-containing layer 44a2. Although the curing-type resin layer-containing bead-containing transfer foil 44 as

shown in FIG. 17(c) is constituted in a same manner as in the transfer foil as in FIG. 17(b), an interlayer 44a4 is provided between the bead-containing layer 44a2 and the adhesive layer 44a3. Although the curing-type resin layer-containing bead-containing transfer foil 44 as shown in FIG. 17(d) is constituted in a same manner as in the transfer foil as shown in FIG. 17(c), a barrier layer 44a5 is provided between the interlayer 44a4 and the bead-containing layer 44a2. Although a transfer foil as shown in FIG. 17(e) is constituted in a same manner as in the transfer foil as shown in FIG. 17(d), an adhesive layer 44a8 is provided between the resin layer 44a9 and the bead-containing layer 44a2, and the barrier layer 44a5 is omitted.

[0080]

These embodiments of the curing-type resin layer-containing bead-containing transfer foil 44 are each transferred by allowing the curing-type resin layer-containing bead-containing transfer layer 440 to be peeled off from the support 44b.

[0081]

In the transparent protective transfer foil in this embodiment, the transparent protective transfer layer is peeled from the support and transferred. The transparent protective transfer foil is constituted in such a manner that at least a peeling layer, a transparent protective layer and

an adhesive layer are laminated on a support in the stated order, at least a peeling layer, a transparent protective layer, an interlayer and an adhesive layer are laminated on a support in the stated order, or at least a peeling layer, a transparent protective layer, a barrier layer, an interlayer and an adhesive layer are laminated on a support in the stated order. The transparent protective transfer foil is excellent in surface protective properties as well as surface abrasion resistance.

[0082]

Further, the optimally variable element transfer foil is constituted in such a manner that at least a peeling layer, an optically variable device layer and an adhesive layer are laminated on a support in the stated order, at least a peeling layer, an optically variable device layer, an interlayer and an adhesive layer are laminated on a support in the stated order, or at least a peeling layer, an optically variable device layer, a barrier layer, an interlayer and an adhesive layer are laminated on a support in the stated order. The foil is excellent in surface protective properties as well as surface abrasion resistance.

[0083]

Still further, the transparent protective transfer foil as well as the optically variable device transfer foil has a transparent protective layer between the peeling layer and the

optically variable device layer and the transparent protective layer may be a cured transparent protective layer.

[0084]

Even still further, in order to obtain more excellent surface protective properties as well as surface abrasion resistance, the transparent protective layer, which is positioned closer to the surface side of the image recording body than the optically variable device layer, is preferably an ultraviolet radiation-cured layer or an electron ray-cured layer.

[0085]

Furthermore, in order to realize prevention curing for the forgery or the alteration, the optically variable device layer is preferably a hard coat layer or an evaporated layer having an image on an uneven surface. [0086]

Still furthermore, in order to obtain excellent surface abrasion resistance, at least one transparent protective layer is thermally transferred onto an entire surface of a card. [0087]

Even still furthermore, an antistatic agent is preferably incorporated into either the transparent protective transfer foil or the optically variable device transfer foil. Thus, it is possible to prepare cards or sheets which are resistant to dust adhesion.

[8800]

Further, the surface formed by the transfer of a layer is preferably subjected to an adhesion treatment so that the transfer foil subsequently transferred is readily adhered.

[0089]

In the transfer foil according to the present invention, one antistatic layer is preferably provided in at least one of a peeling layer, a transparent protective layer, an optically variable device layer, a barrier layer, an interlayer and an adhesive layer. The antistatic layer in the transfer foil contains an anionic high molecular compound and/or an electrically conductive particle, which exhibits excellent antistatic properties.

[0090]

As for such anionic high molecular compounds as described above, polymers each having a carboxyl group, a sulfonic acid group or a hydroxyl group, for example, polyacrylic acid, polymethacrylic acid, vinyl chloride, maleic acid mono(2-ethylhexyl) copolymer, polystyrene sulfonic acid, polyvinyl alcohol, cellulose, hydroxymethyl cellulose and modified compounds thereof are mentioned. Those in which a portion of a functional group or an entire functional group corresponding to each of them is modified to an alkali metal salt, an alkali earth metal salt, a transition metal salt or the like are also mentioned. Of these compounds, preferred

are the alkali or alkali earth metal salt of each of polyacrylic acid, polymethacrylic acid and polystyrene sulfonic acid. Particularly, a sodium salt of polystyrene sulfonic acid is preferred from the viewpoint of compatibility with other resins to be described below, antistatic properties, solubility and viscosity in a state of solution.

[0091]

Further, as for such electrically conductive particles, oxides of various types of metals such as ZnO, TiO2, SnO2, Al2 O<sub>3</sub>, In<sub>2</sub>O<sub>3</sub>, MgO, CoO, CuO, Cu<sub>2</sub>O, SrO, BaO, BaO<sub>2</sub>, PbO, PbO<sub>2</sub>, MnO<sub>3</sub>, SiO<sub>2</sub>, ZrO<sub>2</sub>, Ag<sub>2</sub>O, Y<sub>2</sub>O<sub>3</sub>, BiO<sub>3</sub>, La<sub>2</sub>O<sub>3</sub>, Ti<sub>2</sub>O<sub>3</sub>, Sb<sub>2</sub>O<sub>3</sub>, Sb<sub>2</sub>O<sub>5</sub>, K<sub>2</sub>Ti<sub>6</sub> O<sub>13</sub>,  $NaCaP_2O_{18}$  and  $MgB_2O_5$ , sulfides thereof such as CuS and ZnS, carbides thereof such as SiC, TiC, ZrC, VC, NbC and WoC, nitrides thereof such as Si<sub>3</sub>N<sub>4</sub>, Tin, ZrN, VN, NbN, TaN and Cr<sub>2</sub>N, borides thereof such as TiB2, ZrB2, NbB2, TaB2, CrB, MoB, WB and LaB6, silicides thereof such as TiSi2, ZrSi2, NbSi2, TaSi2, CrSi<sub>2</sub>, MoSi<sub>2</sub> and WSi<sub>2</sub>, metal salts such as BaCO<sub>3</sub>, CaCO<sub>3</sub>, SrCO<sub>3</sub>, BaSO<sub>4</sub> and CaSO<sub>4</sub>, composites such as  $Si_3N_4$ -SiC and  $9Ai_2O_3$ - $2B_2O_3$ , articles doped with any one of these compounds and the like are mentioned. Among these compounds and articles, ZnO, TiO2,  $SnO_2$ ,  $In_2O_3$ ,  $ZrO_2$ ,  $Y_2O_3$ ,  $Sb_2O_5$ , CuS, ZnS, TiC, WC, Tin,  $TiB_2$ ,  $ZrB_2$ ,  $MoSi_2$ ,  $WSi_2$ ,  $CaCO_3$ ,  $BaSO_4$  and the like are preferred. compounds may be employed each individually or in combination of two types or more.

[0092]

In the present invention, the electrically conductive particles are preferably a mixture of at least two types of having different particle particles shapes. Entire electrically conductive particles are preferably made up of some of particles having an average particle diameter of from  $0.01\ \text{to}\ 0.5\ \mu\text{m}$  and the remaining particles having an average particle diameter of 0.5 to 3  $\mu m$ , and a preferred ratio is such that the former is from 50 to 95% by weight and the latter is from 5 to 50% by weight. When an antistatic layer is formed by employing only particles having an average particle diameter of from 0.01 to 0.5  $\mu m_{\textrm{\tiny J}}$  the resulting layer is effective in minimizing static charge as well as transparent spots. However, the following problems occasionally occur: plurality of sheets are fed, stable running is not carried out, and adhesion to a sticky surface of an image receiving layer immediately after preparing an image recording body is not minimized. Further, when the antistatic layer is formed by employing only particles having an average particle diameter of from 0.5 to 3  $\mu m_{\star}$  the following effects were obtained: the static charge was minimized, feeding of a plurality of sheets was minimized, stable running was carried out, and adhesion to the sticky surface of the image receiving layer immediately after preparing the image recording body was minimized. However, transparent spots were occasionally formed.

[0093]

As described above, when two types of electrically conductive particles are employed in the antistatic layer, they are effective not only in minimizing the static charge but also in minimizing the feeding of a plurality of sheets, stabilizing running, minimizing transparent spots, and minimizing adhesion to the sticky surface of the image receiving layer immediately after preparing the image recording body.

[0094]

Further, a content of the anionic high polymer compound and/or the electrically conductive particle in an antistatic layer forming composition is preferably from 5 to 80% by weight and, more preferably, from 10 to 60% by weight on the basis of the entire weight.

In the present invention, the antistatic layer is formed by blending the anionic high polymer compound and/or the electrically conductive particle into a resin, or by dispersing it into the resin.

[0096]

[0095]

Resins forming the antistatic layer are not particularly limited, and various types of resins such as known binder resins may be employed. Representative examples of binder resins include methyl polymethacrylate-based acrylic resins, styrene-based resins such as polystyrene, vinyl chloride-based resins such as polyvinyl chloride, vinylidene

chloride-based resins such as polyvinylidene chloride, polyester-based resins such as polyethylene terephthalate, cellulose-based resins such as cellulose acetate, polyvinyl acetal-based resins such as polyvinyl butyral, epoxy-based resins, amide-based resins, urethane-based resins, melamine-based resins, alkyd-based resins, phenol-based resins, fluorine-based resins, silicon-based resins, polycarbonate, polyvinyl alcohol, casein and gelatin. Further, resins such as ionizing radiation-curable resins or thermosetting resins, which are capable of being cured by ionizing radiation or heat after the antistatic layer is formed, may be employed in combinations.

An actinic ray-curable resin layer is provided by employing the transparent protective transfer foil 64 as shown in FIG. 14, the optically variable device transfer foil 43 as shown in FIG. 15, the resin transfer foil 66 as shown in FIG. 16 and the curable resin layer-containing bead-containing transfer foil 44 either each individually or in combinations. [0098]

[0097]

As described above, the transparent protective layer as well as the optically variable device layer are provided on the image recording layer and, then, an actinic ray-cured layer having a breaking elongation in the range of from 5 to 90% is provided thereon. Further, an actinic ray-cured layer having

a static surface friction coefficient of from 0.01 to 0.5 is provided. By providing the actinic ray-cured layer having the specified breaking elongation or static surface friction coefficient, scratch resistant is enhanced, and abrasion as well as wrinkling may be minimized.

[0099]

The actinic ray-cured layer contains a non-curable resin, or contains an unsaturated group-containing resin having an average molecular weight of from 5,000 to 50,000. By such incorporation, it is possible to obtain the actinic ray-cured layer having a breaking elongation in the range of from 5 to 90%.

[0100]

Further, the actinic ray-cured layer contains Si and F compounds which include Si and F compounds each having an unsaturated group. Further, the actinic ray-cured layer contains a coupling agent. As a result, the actinic ray-cured layer having a static surface friction coefficient in the range of from 0.01 to 0.5 can be obtained.

[0101]

As described above, the actinic ray-cured layer is provided. Thereafter, the resulting layer is subjected to exposure of actinic ray. Thus, a cured layer is formed, to thereby produce a card.

[0102]

Further, the transfer foil according to the present invention, as shown in FIG. 18(a), contains at least a resin layer 36 having a breaking elongation in the range of from 5 to 90% on the side having releasability of a releasable support 34, and further contains at least a resin layer 36 having a static surface friction coefficient in the range of from 0.01 to 0.5. Still further, the resin layer 36 includes at least one actinic ray-curable resin layer. Even still further, the resin layer 36 may contain a non-curable resin. Furthermore, the resin layer 36 contains an unsaturated group-containing resin having an average molecular weight of from 5,000 to 50,000. Still furthermore, the resin layer 36 contains Si and F compounds which include unsaturated group-containing Si and compounds. Even still furthermore, the resin layer 36 contains a coupling agent.

[0103]

Further, after the resin layer is formed, as shown in FIG. 18(b), an interlayer 36a and an adhesive layer 36b are successively provided. Alternatively, after the resin layer is formed, as shown in FIG. 18(c), a bead-containing layer 36c, the interlayer 36a and the adhesive layer 36b are successively provided.

[0104]

As shown in FIG. 18(b) as well as in FIG. 18(c), the transfer foil is transferred onto an adherend 37, to thereby

prepare a card. In this case, the transfer foil may be transferred onto the adherend 37 in which a transparent protective layer and/or an optically variable device layer has previously been transferred onto the image recording layer. [0105]

Further, as shown in FIG. 18(d), in the production method for the transfer foil according to the present invention, after applying an actinic ray-cured layer 36d to the side having releasability of the releasable support 34, exposure is carried out by employing actinic ray, to thereby form a cured layer. Thereafter, the transfer foil may be produced by successively providing the interlayer 36a and the adhesive layer 36b. [0106]

Still further, as shown in FIG. 18(e), after applying the actinic ray-cured layer 36d, actinic ray exposure is carried out, to thereby form a cured layer. Thereafter, the transfer foil may be produced by successively providing a bead-containing layer 36e, the interlayer 36a and the adhesive layer 36b.

[0107]

Even still further, as shown in FIG. 18(f), after applying the actinic ray-cured layer 36d on the side having releasability of the releasable support, actinic ray exposure is carried out, to thereby form a cured layer. Thereafter, an adhesive layer 36g, the bead-containing layer 36e, the

interlayer 36a and the adhesive layer 36b are successively provided.

[0108]

The transfer foil produced by employing the transfer foil production method, as described above, is transferred onto the adherend 37 to make it possible to form a card. Further, the transfer foil produced by employing the transfer foil production method is transferred onto an adherent in which an optically variable device layer has previously been transferred to make it possible to form the card.

[0109]

Furthermore, the interlayer contains at least a polyvinyl butyral resin having a polymerization degree of 1,000 or more, or a thermosetting resin-based on polybutyral, and the adhesive layer contains at least a urethane-modified ethylene ethylacrylate copolymer and a polyacrylic acid ester copolymer.

[0110]

In a manner as described above, security such as prevention of the forgery or the alteration can be enhanced, and, at the same time, abrasion as well as wrinkling can be minimized, compared with a conventional method.

[0111]

The card material according to the present invention will now be described below.

[0112]

The support onto which the present device can be adhered are made of such base materials as papers, e.g., paper, polypropylene, polystyrene, or synthetic papers prepared by laminating these with paper; plastic films, e.g., transparent or white polyethylene terephthalate base film, polyethylene terenaphthalate base film and vinyl chloride base film; various types of metallic or ceramic films and the like. IC memory, optical memory, magnetic memory or the like may be built in the base body which may be either of single layer or a composite film of aforementioned films. Thickness of the base body is ordinarily from about 100 to 1,000  $\mu$ m and, preferably, from 200 to 700  $\mu$ m. On a reverse surface, a writing layer may be provided.

[0113]

Next, the image recording layer according to the present invention will be described.

[0114]

The image element according to the present invention is formed in such a manner that a protective layer is formed by employing a thermal transfer sheet on the image surface on an information-carrying layer on which at least one selected from an identification image such as a portrait and the like, an attribute information image, and format print is provided or on a print surface.

[0115]

Listed as methods to form identification represented by a portrait, are a silver salt photographic method which is advantageous for formation of multi-level images, a fusion thermal transfer recording method, an ink jet method, and a sublimation type thermal transfer recording method. In recent years, as represented by the driver's license, the latter is ordinarily employed. The attribute information means names, addresses, birth date, qualifications and the like, and is recorded as text information, ordinarily employing a fusion type thermal transfer recording method. The format printing is carried out on a base body or an image receiving layer employed for a sublimation type thermal recording method, employing printing methods such as resin letterpress printing, planographic printing, silk screen printing. Appropriately selected as a pattern layer are printed matter, holograms, bar codes, matted patterns, fine patterns, ground patterns, concave and convex patterns, and the like. The pattern layer contains visible light absorbing colorants, UV absorbing materials, infrared ray absorbing materials, fluorescent whitening materials, an evaporated metal layer, an evaporated glass layer and the like. The interlayer exhibits a cushioning function for absorbing the unevenness of a body to which transfer is carried out, and a resin curing function for the purpose of protecting the

pattern layer after transfer, and a light-cured resin layer and a thermally cured resin layer are added.
[0116]

Transfer of the transfer foil to the transfer material is ordinarily carried out by employing means such as a thermal head, a heated roller and a hot stamp machine which can apply pressure while heating.

[0117]

The image receiving layer according to the present invention, which is a layer possessing identification information or bibliographical information, is preferably constituted in such a manner that at least an identification information image can be formed on its surface by employing sublimable dyes and the like. Its constitution is not particularly limited, so long as it is capable of accepting sublimable dyes which are thermally diffused from the ink layer in the sublimation type thermal transfer ink sheet, and it basically contains a binder and, optionally, various types of additives. Thickness of the image receiving ordinarily from 1 to 50  $\mu m$  and, preferably, from about 2 to about 20  $\mu m$ .

[0118]

Employed as binders for the image receiving layer may be various types of resins such as vinyl chloride-based resins, polyester-based resins, polycarbonate-based resins, acrylic

resins and polyvinyl acetal-based resins. However, from the viewpoint of image retention properties and the like, preferred are polyvinyl acetal-based resins such as polyvinyl acetoacetal resins, polyvinyl butyral resins and polyvinyl formal resins, or vinyl chloride-based resins such as polyvinyl chloride resins and vinyl chloride copolymers. In addition to these compounds, polyester-based resins may preferably be employed as an image receiving layer for sublimation type thermal transfer.

[0119]

As additives, releasing agents, antioxidants, UV absorbers, light stabilizers, fillers, and pigments may be incorporated into the image receiving layer. Further, incorporated as sensitizers, may be plasticizers, heat solvents, and the like into the same. Listed as releasing agents are silicone oil (including those called silicone resins); solid waxes such as polyethylene wax, polypropylene wax, amide wax and Teflon powder; fluorine-based and phosphoric acid-based surfactants. Of these agents, silicone oil is preferred.

[0120]

Cited as antioxidants may be those as described in JP-A Nos. 59-182785, 60-130735, 1-127387 and the like, and compounds known in the art, which improve image durability in photographic or other image recording materials.

[0121]

Cited as UV absorbers as well as light stabilizers may be those as described in JP-A Nos. 59-158287, 63-74686 and 63-145089, and other compounds known in the art which improve image durability in photographic or other image recording materials.

[0122]

Listed as fillers may be inorganic particles such as silica gel, calcium carbonate, titanium oxide, acid clay, active clay and alumina, and organic particles such as fluorine resin particles, guanamine resin particles, acrylic resin particles and silicone resin particles.

[0123]

Listed as representative pigments may be titanium white, calcium carbonate, zinc oxide, barium sulfate, silica, talc, clay, kaolin, active clay, acid clay and the like.
[0124]

In addition, in order to effectively minimize fusion between the image receiving layer and the ink layer of an ink sheet for thermal transfer recording, a peeling layer containing releasing agents may be applied onto the surface of the image receiving layer. Thickness of the peeling layer is ordinarily from 0.03 to 2.0  $\mu m$ . Further, either a cushion layer or a barrier layer may be provided between the support for ID cards and the image receiving layer. Other than these

layers, image recording layers described in JP-A Nos. 07-117367, 08-324117, 08-282196, 09-44065 and the like may also be employed. However, the present invention is not limited to these examples.

[0125]

In the sublimation image forming method, an ink sheet for sublimation type thermal transfer recording is employed. The sheet for sublimation type thermal transfer recording can be constituted with a support and a sublimable dye-containing ink layer formed thereon.

[0126]

The supports are not particularly limited, and those conventionally known in the art may be employed, so long as sufficient dimensional stability as well as heat resistance during recording with a thermal head is exhibited.
[0127]

The sublimable dye-containing ink layer basically contains sublimable dyes as well as binders. Listed as sublimable dyes may be cyan dyes, magenta dyes, and yellow dyes. Listed as cyan dyes naphthoquinone-based are anthraquinone-based dyes, azomethine-based dyes and the like as described in JP-A Nos. 59-78896, 59-227948, 60-24966, 60-53563, 60-130735, 60-131292, 60-239289, 61-19396, 61-22993, 61-31292, 61-31467, 61-35994, 61-49893, 61-148269, 62-191191, 63-91288, 63-91287, 63-290793 and the like.

[0128]

Listed as magenta dyes are anthraquinone-based dyes, azo dyes, azomethine-based dyes and the like as described in JP-A Nos. 59-78896, 60-30392, 60-30394, 60-253595, 61-262190, 63-5992, 63-205288, 64-159, 64-63194 and the like. Listed as yellow dyes are methane-based dyes, azo-based dyes, quinophthalone-based dyes, and anthraisothiazole-based dyes as described in JP-A Nos. 59-78896, 60-27594, 60-31560, 60-53565, 61-12394, 63-122594 and the like. [0129]

Further, particularly preferred sublimable dyes include azomethine dyes which are obtained through coupling reaction of compounds having a open chain type or closed chain type active methylene group with oxidized p-phenylene diamine derivatives or oxidized p-aminophenol derivatives, and indoaniline dyes which are obtained through coupling reaction of phenol or naphthol derivatives with oxidized p-phenylene diamine derivatives or oxidized p-aminophenol derivatives. [0130]

Further, when metal ion-containing compounds are incorporated into the image receiving layer, sublimable dyes, which react with the metal-containing compounds to form chelates, are preferably incorporated into an sublimable dye-containing ink layer. Listed as such sublimable dyes capable of forming chelates may be cyan dyes, magenta dyes,

and yellow dyes capable of forming bidentate chelates, which are described in, for example, JP-A Nos. 59-78893 and 59-109349, and Japanese Patent Application Nos. 2-213303, 2-214719 and 2-203742. Preferred sublimable dyes capable of forming a chelate may be represented by the general formula described below:

[0131]

X1-N=N-X2-G

wherein X1 represents an aromatic carbon ring in which one ring contains 5 to 7 atoms, or an atomic group necessary for completing a heterocyclic ring, and at least one in the position adjacent to a carbon atom bonded to an azo bond is a nitrogen atom or a carbon atom substituted with a chelating group;

X2 represents an aromatic heterocyclic ring or an aromatic carbon ring in which at least one ring contains 5 to 7 atoms; and

G represents a chelating group. [0132]

Sublimable dyes incorporated into the aforementioned sublimable dye-containing ink layer, when an image to be prepared is monochromatic, may be any of yellow dyes, magenta dyes and cyan dyes. Depending on the color of the image to be prepared, at least any two among the three dyes or other sublimable dyes may be incorporated. An amount of sublimable

dyes to be used is ordinarily from 0.1 to 20 g per m² of the support and, preferably, from 0.2 to 5 g per m² of the support. Binders in the ink layer are not particularly limited, and those, which have heretofore been known in the art, may be employed. Further, into the ink layer, suitably incorporated may be various types of additives which have heretofore been known in the art. Ink sheets for sublimation type thermal transfer recording are produced in such a manner that a coating composition used to form the ink layer is prepared by dispersing or dissolving the aforementioned various components in a solvent, and the resulting coating composition is applied onto a surface of the support and subsequently dried. Thickness of the ink layer thus prepared is ordinarily from 0.2 to 10  $\mu$ m and, preferably, from 0.3 to 3  $\mu$ m.

Next, the transfer foil according to the present invention will be described.

[0133]

[0134]

Employed as supports for the transfer foil are heat resistant plastic films made of polyester, polyethylene terephthalate, polyethylene naphthol, an ABS resin, and the like. A peeling layer is provided on one surface thereof as a bottom layer and an adhesive layer is provided as a top layer. If desired, a pattern layer, an interlayer and the like are provided between the peeling layer and the adhesive layer.

Thickness of plastic film supports is ordinarily from 3 to 50  $\mu m$  and, preferably, from 10 to 30  $\mu m$  . [0135]

As for the peeling layer of the transfer foil made of resins such as an acrylic resin having a high glass transition temperature, a polyvinyl acetal resin and a polyvinyl butyral, waxes, various types of silicone oil, fluorine compounds, water-soluble polyvinylpyrrolidone, polyvinyl Si-modified polyvinyl alcohols, a methyl cellulose resin, a hydroxy cellulose resin, silicone resins, paraffin waxes, acryl-modified silicone, polyethylene waxes, resins such as ethylene vinyl acetate and, in addition, polydimethylsiloxane and modified products thereof, for example, oil and resins such polyester-modified silicone, acryl-modified silicone, urethane-modified silicone, alkyd-modified silicone, amino-modified silicone, epoxy-modified silicone, polyether-modified silicone and cured products thereof. Cited as other fluorine-based compounds are fluorinated olefins, perfluorophosphoric acid ester-based compounds. Listed as preferred olefin-based compounds are dispersions of polyethylene, polypropylene and the like and long chain alkyl-based compounds such as polyethyleneimine octadecyl. Some releasing agents, which exhibit low solubility, may be dispersed and employed.

[0136]

Further, it is possible to carry out addition to other polymers in the same manner as in silicone compounds. Other than these compounds, fine particles (having a diameter in the order of micron or submicron) and the like are effective. Thickness thereof is preferably from 0.1 to 2  $\mu m$ . [0137]

Listed as compounds forming the adhesive layer of the transfer foil are thermally adhesive resins such as ethylene vinyl acetate resins, ethylene ethyl acrylate resins, ethylene acrylic acid resins, ionomer resins, polybutadiene resins, acrylic resins, polystyrene resins, polyester resins, olefin resins, urethane resins and adhesion providing agents (for instance, phenol resins, rosin resins, terpene resins and petroleum resins). Copolymers as well as mixtures thereof may be employed. Thickness thereof is preferably from 0.1 to 10  $\mu m$ .

[0138]

Specifically, urethane-modified ethylene ethyl acrylate copolymers are commercially available such as Hitech S-6254, S-6254B and S-3129, manufactured by Toho Kagaku Kogyo Co., Ltd., and polyacrylic acid ester copolymers are commercially available as Jurimar AT-210, AT-510, and AT-613, manufactured by Nihon Junyaku Co., Ltd.; Plussize L-201, SR-102, SR-103, J-4, and others, manufactured by Ogo Kagaku Kogyo Co. The weight ratio of urethane-modified ethylene ethyl acrylate

copolymers to polyacrylic acid ester copolymers is preferably from 9:1 to 2:8. Thickness of the adhesive layer is preferably from 0.1 to 1.0  $\mu m\,.$ 

[0139]

The interlayer in the transfer foil preferably contains at least two layers. When required, a primer layer as well as a barrier layer may be provided as the interlayer, and adhesion between layers may further be enhanced. as specific compounds are thermoplastic resins constituted with block polymers of polystyrene and polyolefin and polyvinyl butyral. Polyvinyl butyral resins each polymerization degree of 1,000 or more, in the interlayer according to the present invention, are commercially available such as Esurekku BH-3, BX-1, BX-2, BX-5, BX-55 and BH-S manufactured by Sekisui Kagaku Kogyo Co., Ltd., and Denka Butyral #4000-2, #5000-A and #6000-EP manufactured by Denki Kagaku Kogyo Co., Ltd., and the like. The polymerization degree before thermosetting of polybutyral thermosetting ' resins in the interlayer is not particularly limited, and resins which each have a lower polymerization degree may be employed. Isocyanate curing agents, epoxy curing agents and the like may be employed for thermosetting, and thermosetting conditions are preferably from 50 to 90°C for from 1 to 24 hours. Thickness of the interlayer is preferably from 0.1 to 1.0  $\mu m$ . [0140]

Next, the bead-containing layer containing beads according to the present invention will be described.
[0141]

The bead-containing layer containing beads according to the present invention reconstitutes incident light by adding a phase difference to a portion of the incident light, emphasizes a light component of a specific wavelength region by interference to return a colored light different in color tone from the incident light to an incoming direction of the incident light; and contains a reflection substrate and transparent beads aligned thereon. The bead-containing layer containing beads is formed of a reflection substrate provided with a resin layer thereon, and many beads made of, e.g., glass and having a bead diameter of from 10 to 60  $\mu m$ , preferably, from 15 to 40  $\mu m\text{,}$  are further aligned on the surface side thereof. A light refractive index of the bead is preferably from 1.6 to 2.1, more preferably, from 1.7 to 2.0. The incident light incoming from outside proceeds into the beads, at least a part of the light being reflected by the reflection substrate through the transparent beads and resin layer, to return to the beads again and proceeds to outside. Because the outwardly extruding surfaces of the beads are spherical, a similar function is generated regardless of minor variations in the incident angle and the reflected light can be returned to the incident direction.

[0142]

Next, the reflection layer according to the present invention is described.

[0143]

[0144]

The reflection layer according to the present invention is selected from at least a metal thin film, a metal oxide thin film, a light interference substance and a light diffraction layer. The reflection layer is preferably formed by printing, in arbitrary tint patterns, a paint containing powder such as interference substances, metal oxides and mica, capable of each exhibiting an interference color.

Examples of metal oxides include titanium dioxide, ferric oxide, titanium lower-order oxide, zirconium oxide, silicon oxide, aluminum oxide, cobalt oxide, nickel oxide and cobalt titanate; complex oxides such as Li<sub>2</sub>CoTi<sub>3</sub>O<sub>8</sub> and KNiTiO<sub>x</sub>; and mixed substances of these metal oxides, however, are not limited to these metal oxides, so long as they are metal oxides capable of exhibiting an interference color. As an interference substance layer, a metal film, having an interference color obtained by oxidizing the surface thereof, can be used. Methods to prepare these metal films include a method in which films of, for example, aluminum metal, titanium metal and stainless steel are anodically oxidized; a method in which a metal oxide capable of exhibiting interference color is

prepared by a sol-gel method and coated; a method in which an alkoxide of metal capable of exhibiting interference color is coated on a metal film and thermally decomposed; and evaporation process such as a CVD and a PVD.

[0145]

Next, the pearl pigment according to the present invention will be described.

[0146]

The pearl pigment is used for increasing a reflection light amount at the time of recording. The reflection light amount necessary at the time of recording can be obtained, so long as it has a high reflection ratio of 30% or more in an entire visible light region. This pearl pigment is a pigment having pearlescence and is a crystal of a carbonate, arsenate or the like into which light is incident and performs a regular multiple reflection, to thereby exhibit the pearlescence.
[0147]

The pearl pigment to be used in the present invention is formed such that mica is coated by a mono layer or two layers with a metal oxide which gives reflection (high iris reflection), is transparent in a visible region and has a refractive property of 2 or more; examples of such metal oxides include Sb<sub>2</sub>S<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, PbO, ZnSe, CdS, Bi<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, PbCl<sub>2</sub>, CeO<sub>2</sub>, Ta<sub>2</sub>O<sub>5</sub>, ZnS, ZnO, CdO, Nd<sub>2</sub>O<sub>3</sub>, Sb<sub>2</sub>O<sub>3</sub>, SiO and InO<sub>3</sub>.

When mica and a metal oxide film are combined with each other, since a difference between refractive indices thereof is larger than 0.4, a reflection amount of incident white light is large and, at the same time, birefringence is generated at an interface between mica and the metal oxide film and, as a result, the pearl pigment comes to have a high iris reflection property which works for more effectively promoting a color change effect. On this occasion, by controlling a film thickness of the metal oxide film which coats mica, the pearl pigment having an arbitrary color tone and the high iris reflection property can be prepared and the film thickness thereof is preferably in the range of from 10 to 10000 angstroms and, desirably, in the range of from 200 to 2000 angstroms, since the pearl pigment comes to have a high iris reflection property in a visible range in the stated range. As for such pearl pigments as being available in the market, "Iriodin" (trade name; manufactured by Merck Co.) is mentioned. "Iriodin" is an inorganic pearl pigment in which a surface of natural mica is coated with a metal oxide having a high refractive index such as titanium oxide and iron oxide. Light reflected at the interface between a layer of titanium oxide having a high refractive index and mica having a low refractive index together with a neighboring medium emits pearlescence. This "Iriodin" can emphasize a specified color in iris colors by changing film thickness of titanium oxide to be coated. Such

pearl pigment as described above can be used after being made into ink form by being added with a colored pigment, a vehicle and an additive.

[0149]

The pearl pigment to be used in the printed layer may be those as described above, so long as it has a different interference property from that of the pearl pigment to be used in the reflection layer. Other than these types, a gold type or a silver type can be used. Further, an optically variable device layer of, for example, hologram may be provided thereto.
[0150]

In the present invention, an actinic ray-cured layer having a breaking elongation of 5 to 90% is provided, and an actinic ray-cured layer having a static surface friction coefficient of 0.01 to 0.5 is also provided. As a result, scratch resistance is enhanced and excellent surface protection is exhibited.

[Measurement Method of Breaking Elongation]

The breaking elongation (in %) in the present invention was measured as described below.

A resin layer was left untouched at ambient conditions of 23°C 55% RH for at least one hour. Thereafter, the resulting layer was measured by employing a Tensiron Multipurpose Tester RTA-100 of Orienteku Co., Ltd., while carrying out data processing by employing Tensiron Multifunctional Type Data

Process Type MP-100/200S Ver. 44. The resin was fixed by employing an air-chuck method. It is possible to select each of ranges as follows: 5 to 100 mm/minute for the cross head speed; 5 to 100% for the RANGE; and 0.1 to 500 kg for the load. In the present invention, evaluation was carried out at a cross head speed of 30 mm/minute, a RANGE of 20% and a load of 100 kg.

[0151]

Further, when the breaking elongation of resins or actinic ray-curable resins is measured, it is difficult to prepare a single resin film made of only the actinic ray-cured layer. Therefore, a 5 µm thick resin layer is formed on a 12 µm thick PET. The actinic ray-cured layer was conveyed two times at a 5 m of CS under a mercury lamp having an exposure energy of 160 W/cm, to thereby cure the layer. Measurements were carried out in such a manner that a 1 cm wide sample was fixed by employing an air chuck and pulling tests were carried out.

[0152]

The breaking elongation was obtained based on the breaking elongation point at which during pulling resins or actinic ray-curable resins, breaking or cracking occurred.

[Measurement Method of Static Friction Coefficient]

Static friction coefficient ( $\mu s$ ) can be obtained based on the same principle as employed in Friction Coefficient Test

Method defined in JIS K7125. After a resin is left untouched at conditions of 25°C and 60% RH for at least one hour, a constant load (contact force Fp, for example, of 50 to 200 g) is applied to a steel ball (for instance, having a diameter of 0.5 to 5 mm), which is allowed to slide on the surface of the resin layer at a constant speed (for example, 2 to 100 cm/minute). During sliding, the first maximum load (a static friction force) (Fs) is measured and the static friction coefficient is obtained by employing the following numeral 1:

μs=Fs/Fp,

wherein  $\mu s$  represents the static friction coefficient; Fs represents the static friction force (kgf); and Fp represents the contact force N (kgf).

In the present invention, measurements were carried out by employing a Surface Properties Measurement Instrument Heidon (Type 14DR) manufactured by Shinto Kagaku Co., Ltd., at the conditions of a 100 g load, as well as a speed of 600 mm/minute. Further, measurements were carried out by employing a load converter 100 gf and a ball pressure element (a 5 mm chrome ball) as the measurement devices.

As for actinic rays, anything generating an actinic electromagnetic wave to a polymerization initiator can be employed. For example, a laser, a light emitting diode, a xenon

flash lamp, a halogen lamp, a carbon arc lamp, a metal halide lamp, a tungsten lamp, a mercury lamp and an electrodeless lamp can be mentioned. As examples of preferable light sources, a xenon lamp, a halogen lamp, a carbon arc lamp, a metal halide lamp, a tungsten lamp and a mercury lamp are mentioned. Energy to be added in this instance can optionally be employed by so selecting as adjusting exposure distance, time and strength in accordance with a type of polymerization initiator. The actinic ray can increase polymerization rate by intercepting air by a method of nitrogen substitution, reduced pressure or the like, depending on cases.

[0155]

When a laser is employed as a light source, it is easy to narrow down exposure spot to a minute size and, therefore, it is possible to form an image of high resolution. As laser light sources, any one of an Ar laser, a He-Ne gas laser, a YAG laser and a semiconductor laser can suitably be employed. Further, when photo-curing is done using an actinic ray, reduced pressure and means to stabilize the photo-curing in nitrogen gas stream or the like may be employed. As for means which can add thermal energy to the actinic ray, any one of an oven, a heat-roll, a hot stamp, a thermal head, laser light, infrared ray flash, a thermal pen and the like may timely be selected and, then, used.

[Coupling Agent]

The coupling agent according to the present invention represents a coupling agent of, for example, silicone type, aluminum type or titanium type, and those as described in JP-A Nos. 2-4258 and 4-161957 can be mentioned. When the coupling agent is added to a resin layer or a cured layer, it is added, based on the entire solid content, in an amount of from 0.1 to 30% by weight and, preferably, in the range of from 1 to 20% by weight. The coupling agent can also be used as a surface treatment layer of the resin layer or the cured layer. [Resin having Preferable Breaking Elongation]

Any resin having a breaking elongation of from 5 to 90% may be employed. Specifically, those resins as described in JP-A No. 11-184270 may be employed.

[Resin having Preferable Static Frictional Coefficient]

Any resin layer having a static frictional coefficient in the range of from 0.01 to 0.5 may be employed. Examples of such resins include a silicone type resin, a fluorine type resin, a polyethylene wax and an amide wax. It is particularly preferred to employ a cured layer formed by an actinic ray in view of a manufacturing process, a manufacturing cost and a good final quality (specifically, refer to Japanese Patent Application No. 11-184270).

[Actinic Ray Cured Layer]

An actinic ray-curable resin layer according to the present invention contains a material having an addition

polymerizable property or an open chain polymerizable property. An addition polymerizable compound includes a radically polymerizable compound such as photopolymerizable material employing a photo(or thermo)polymerizable composition as described in, for example, JP-A No. 7-159983, JP-B No. 7-31399, JP-A Nos. 8-224982 and 10-863, Japanese Patent Application Nos. 7-231444, 7-231444 and 11-184270. The photocurable material οÍ cationic polymerization is known the addition as polymerization compound, and a photocurable material of photo-cation polymerization sensitized in a longer wavelength than a visible ray has recently been disclosed, for example, in JP-A Nos. 6-43633 and 8-324137. Both may be employed for the object according to the present invention. One or more layers cured by the actinic ray are provided in the layer arrangement, and at least one layer satisfies the breaking elongation or static frictional coefficient as stipulated by the present invention. It is preferable that an outermost layer in the card satisfies the frictional coefficient stipulated by the present invention.

[0156]

[Initiator for Actinic Ray Curing]

A radical polymerization initiator is given for another compound generating a radical or an acid which may simultaneously be employed with a radically polymerizable composition containing a specified compound according to the

present invention. Further, the specified compound according to the present invention generates a radical (such as Cl) by light or heat, and the radical withdraws a proton in the layer and generates an acid (such as HCl), to thereby conduct polymerization. At this stage, the polymerization initiator mentioned below, which may be employed in combination, generates a radical or an acid by light or heat, to thereby conduct the polymerization (specifically, refer to Japanese Patent Application 11-184270). No. The radically polymerizable compound or the cationic polymerization type is preferably employed. Further, the radically polymerizable compound is more preferably employed.

1) Cationic Polymerization Type Photocurable resin Cationic Polymerizable Compound

Examples of epoxy type UV ray-curable prepolymers or monomers which are each such type (predominantly epoxy type) as causes polymerization based on cationic polymerization include prepolymers each containing two or more epoxy groups in a molecule. Examples of the prepolymers include alicyclic epoxides, polyglycidyl esters of polybasic acid, polyglycidyl ethers of polyalcohol, polyglycidyl ethers of polyoxyalkylene glycol, polyclycidyl glycol ethers of aromatic polyols, hydrogen adduct compounds of polyglycidyl ethers of aromatic polyols, urethane polyepoxy compounds and epoxy-modified polybutadienes. These prepolymers may be employed solely or

by mixing two or more compounds in combination. [0157]

2) Radical Polymerization Type Photocurable Resin Radically polymerizable Compound

Radically polymerizable compounds to be contained in radically polymerizable compositions include ordinary photopolymerizable compounds and thermopolymerizable compounds. Such radically polymerizable compound is a compound having an ethylenic unsaturated bond capable of performing radical polymerization, and any compound having at least one ethylenic unsaturated bond capable of performing radical polymerization in the molecule. Such compounds include those as each having a chemical form of monomer, oligomer, polymer and the like. The radically polymerizable compound may be employed solely or two or more in arbitrary ratios for improving targeted characteristics.

[0158]

Examples of compounds each having the ethylenic unsaturated bond capable of performing radical polymerization include unsaturated carboxylic acids such as acrylic acid, methacrylic acid, itaconic acid, crotonic acid, isocrotonic acid, maleic acid and salts, esters, urethanes, amides, anhydrides thereof, acrylonitrile, styrene and, further, various types of radically polymerizable compounds such as unsaturated polyesters, unsaturated

polyamides and unsaturated urethanes. Specifically mentioned are acrylic acid derivatives such as 2-ethylhexyl acrylate, 2-hydroxyethyl acrylate, butoxyethyl acrylate, carbitol acrylate, cyclohexyl acrylate, tetrahydrofurfuryl acrylate, benzyl acrylate, bis(4-acryloxy polyethoxyphenyl)propane, neopentyl glycol diacrylate, 1,6-hexanediol diacrylate, ethylene glycol diacrylate, diethylene glycol diacrylate, triethylene glycol diacrylate, tetraethylene glycol diacrylate, polyethylene glycol diacrylate, polypropylene glycol diacrylate, pentaerythritol triacrylate, pentaerythritol tetraacrylate, dipentaerythritol tetraacrylate, trimethylolpropane triacrylate, tetramethylolmethane tetraacrylate, oligoester acrylate, N-methylol acrylamide, diacetone acrylamide and acrylate; methacrylic acid derivatives such methacrylate, n-butyl methacrylate, 2-ethylhexyl methacrylate, lauryl methacrylate, allyl methacrylate, glycidyl methacrylate, benzyl methacrylate, dimethylaminomethyl methacrylate, 1,6-hexane diol dimethacrylate, ethylene glycol dimethacrylate, triethylene glycol dimethacrylate, polyethylene glycol dimethacrylate, polypropylene glycol dimethacrylate, trimethylol ethane trimethacrylate, trimethylol propane trimethacrylate and 2,2-bis(4-methacryloxy poly ethoxyphenyl) propane; and derivatives of allyl compounds such as allyl glycidyl ether,

diallyl phthalate and triallyl trimellitate. Further, radically polymerizable or specifically, crosslinkable monomers, oligomers and polymers available in the market or known in the art which are described in any one of "Crosslinking Agent Handbook" (edited by Shinzo Yamashita, published by Taiseisha in 1981), "UV/EB Curing Handbook" (Material Edition) (edited by Seishi Kato, published by Polymer Publishing Society in 1985), "UV/EB Curing Technical Application and Market", p. 79 (edited by Radtech Kenkyukai, published by CMC in 1989), "Polyester resin handbook" (edited by in Eiichiro Takiyama, published by The Nikkan Kogyo Shimbun Ltd. in 1988) can be employed. A content of the radically polymerizable compound in the radically polymerizable composition is preferably from 1 to 97% by weight and, more preferably, from 30 to 95% by weight. [Additives to Actinic Ray-Curable Resin] Sensitizer

The photosensitive composition according to the present invention can be made to a polymerizable composition having extremely high sensitivity by forming a composition in combination with various types of sensitizers and, then, enhancing activity to light in the region of from ultraviolet to near-infrared. Specific examples of the sensitizers according to the present invention include unsaturated ketones represented by chalcone derivatives, dibenzalacetone and the like; 1,2-diketone derivatives represented by benzyl or

camphor quinine and the like; polymethine dyes such as benzoin derivatives, fluorene derivatives, naphthoquinone derivatives, anthraquinone derivatives, xanthene derivatives, thioxanthene derivatives, xanthone derivatives, thioxanthone derivatives, coumarin derivatives, ketocoumarin derivatives, cyanine derivatives, styryl derivatives, merocyanine derivatives and oxonol derivatives; acridine derivatives; azine derivatives; thiazine derivatives; oxazine derivatives; indoline derivatives; azulene derivatives; azulenium derivatives; squarilium derivatives; porphyrin derivatives; tetraphenylporphyrin derivatives; triaryl methane derivatives; tetrabenzporphyrin derivatives; tetrapyrazino porphyrazine derivatives; phthalocyanine derivatives; tetraazaporphyrazine derivatives; tetraquinoquisariro porphyrazine derivatives; naphthalocyanine derivatives; subphthalocyanine derivatives; pyrylium derivatives; thiopyrylium derivatives; tetraphylline derivatives; annulene derivatives; spiropyran derivatives; spirooxazine derivatives; thiospiropyran derivatives; metal arene complexes; and organic ruthenium complexes. In addition thereto, mentioned are dyes and sensitizers described in any one of "Dye Handbok" (edited by Makoto Okawahara et al., published by Kodansha in 1986), "Kinosei Sikisono Kagaku (Chemistry of Functional Dyes)" (edited by Makoto Okawahara et al., published by CMC in 1981), "Tokushu Kino Zairyo (Special

Functional Material)" (edited by Chuzaburo Ikemori, published by CMC in 1986) and Japanese Patent Application No. 7-108045. Further, mentioned are dyes and sensitizers each having absorption of light in the range of from ultraviolet to near-infrared. Two or more of these compounds may be employed in arbitrary ratios.

[0159]

Polymerization Promoter,

Chain Transfer Agent, etc.

A polymerization promoter or a chain transfer agent may be added to the photosensitive composition according the present invention. Specific examples of such polymerization promoters or chain transfer agents include amines such as N-phenylglycine, triethanolamine and N,N-diethylaniline; thiols as described in USP No. 4,414,312 and JP-A No. 64-13144; disulfides as described in JP-A No. 2-291561; thiones as described in USP No. 3,558,322 and JP-A No. 64-17048; and o-acylthio hydroxamate and N-alkoxypyridine thiones as described in JP-A No. 2-291560.

[0160]

The photosensitive composition according to the invention may be mixed with any one of a dye, an organic or inorganic pigment, an oxygen scavenger such as a phosphine, a phosphonate or a phosphite, a reducing agent, a fog inhibitor, a fading inhibitor, a halation inhibitor, a fluorescent

whitening agent, a surfactant, a coloring agent, an extending agent, a plasticizer, a fire retardant, an oxidation inhibitor, an ultraviolet absorption agent, a foaming agent, a fungicide, an antistatic agent, a magnetic material, any one of other additives which will give various characteristics and a diluting solvent in accordance with purposes.

[0161]

# Polymerization Inhibitor

A polymerization inhibitor may be added to the radically polymerizable composition containing the radically polymerizable compound for the purpose of inhibiting polymerization during reservation of a liquid the photocurable resin layer. Specific examples of thermopolymerization inhibitors which can be added to the radically polymerizable composition include p-methoxyphenol, hydroquinone, alkyl-substituted hydroquinone, catechol, tert-butyl catechol phenothiazine. and These thermopolymerization inhibitors are preferably each added in the range of from 0.001 to 5 parts by weight, on the basis of 100 parts by weight of the compound having a radically polymerizable ethylenic unsaturated bond.

## [Additive to Resin Layer]

In accordance with a need for a non-curing type resin, any one of other arbitrary high molecular weight polymers such as a polyvinylbutyral resin, a polyurethane resin, a polyamide

resin, a polyester resin, an epoxy resin, a novolak resin, styrene, paramethyl styrene, a methacrylate, a vinyl monomer such as an acrylate, a cellulose-based resin, a thermoplastic polyester and a natural resin may simultaneously be used. Further, organic high molecular weight polymers known in the art as disclosed in any one of "New Practical Technology of Photosensitive Resin" (edited by Kiyosi Akamatsu, published by CMC in 1987) and "10188 Chemical Products" pp. 657 to 767 (published by Chemical Daily Co., Ltd. in 1988) may be employed in combination. A content of any one of the organic high molecular weight polymers in the photosensitive composition is preferably in the range of from 1 to 70% by weight and, more preferably, in the range of from 5 to 50% by weight.

[0162]

## Antistatic Agent

As for the antistatic agent, compounds as described in "11290 Chemical Products", pages 875 to 876 (published by Chemical Daily Co., Ltd.) and the like are mentioned in addition to a cationic surfactant, an anionic surfactant, a nonionic surfactant, a polymeric antistatic agent, conductive fine particles and the like.

[0163]

#### Surfactant

Further, a surfactant can be added as an auxiliary agent for allowing the friction coefficient of the photocured layer

to be in the range stipulated by the present invention in the image forming layer of an image forming material according to the present invention. Nonionic surfactants as disclosed in any one of JP-A Nos. 62-251740 and 3-208514 and amphoteric surfactants as described in any one of JP-A Nos. 59-121044 and 4-13149 can be added.

[0164]

Specific examples of nonionic surfactants include sorbitan tristearate, sorbitan monopalmitate, trioleate, monoglyceride stearate and a polyoxyethylene nonylphenyl ether. Further, specific examples of amphoteric surfactants include an alkyl di (aminoethyl) glycine, an alkyl polyaminoethyl glycine hydrochloride, a 2-alkyl-N-carboxyethyl-N-hydroxyethyl imidazolinium betaine and an N-tetradecyl-N, N-betaine type, for example, that having a trade name "Amogen K" (manufactured by Daiichi Kogyo Seiyaku Co., Ltd.).

[0165]

Fluorine Type Surfactant

Fluorine type surfactants each having any one of such structures as described below may be added for the purpose of reducing the friction coefficient stipulated by the present invention. The fluorine type surfactants employed in the present invention are each a polymer made of one, two or more monomers represented by the following general formula (1), (2)

or (3), or a copolymer of the monomer and a monomer capable of being polymerized with this monomer:

$$CH_2=C(R_1)COOARf$$
 (1),

wherein  $R_1$  represents any one of -H, -X (halogen group), an alkyl group and a halogenated alkyl group;

A represents a divalent linking group such as a carbonyl group, an alkylene group, a (poly)oxyalkylene group, a urethane group or a peptide group; and

Rf represents a hydrocarbon group (C: 1 to 30) in which at least one hydrogen atom is substituted with a fluorine atom;

$$CH_2=C(R_1)COO(A)N(R_2)SO_2Rf$$
 (2),

wherein  $R_1$  represents any one of -H, -X (halogen group), an alkyl group and a halogenated alkyl group;

 $R_2$  represents any one of -H, -X (halogen group), an alkyl group and a halogenated alkyl group;

A represents a divalent linking group such as a carbonyl group, an alkylene group, a (poly)oxyalkylene group, a urethane group or a peptide group; and

Rf represents a hydrocarbon group (C: 1 to 30) in which at least one hydrogen atom is substituted with a fluorine atom; and

$$CH_2=C(R_1)Ar(R_2)_1[(A)_mRf]_n$$
 (3),

wherein  $R_1$  represents any one of -H, -X (halogen group), an alkyl group and a halogenated alkyl group;

Ar represents  $-C_6H(_{5-1-n})$ ;

 $R_2$  represents a group which can be substituted with benzene such as -H, -X (halogen group), an alkyl group or a halogenated alkyl group;

A represents a divalent linking group such as a carbonyl group, an alkylene group, a (poly)oxyalkylene group, a urethane group, a peptide group, -O- or -NH-; and

Rf represents a hydrocarbon group (C: 1 to 30) in which at least one hydrogen atom is substituted with a fluorine atom;

1 represents from 0 to 4;

m represents 0 or 1; and

n represents from 1 to 5.

Examples of monomers as described in the above-described general formulae include compounds as described below; however, such monomers are not limited to these compounds.

(1) to (55)

These monomers may each be employed to form a homopolymer copolymer with a or monomer capable of performing polymerization for preparation of a surfactant. As for the monomers which are each capable of performing polymerization with any one of the above mentioned monomers, any compound capable of performing polymerization is permissible. Examples of such monomers include the following compounds but are not limited thereto:

- (1) carboxylic acid vinyl esters such as vinyl acetate, vinyl propionate and vinyl butylate;
- (2) polyoxyalkylene (meta)acrylates such as polyoxyethylene methyl ether (meta)acrylate and polyoxypropylene methyl ether (meta)acrylate;
- (3) ethylenic unsaturated olefins such as ethylene, propylene, isobutylene, butadiene and isoprene;
- (4) styrenes such as styrene,  $\alpha$ -methylstyrene, p-methylstyrene and p-chlorostyrene;
- (5) unsaturated carboxylic acids such as acrylic acid, methacrylic acid, crotonic acid and isocrotonic acid;
- (6) unsaturated aliphatic dicarboxylic acids such as itaconic acid, maleic acid and maleic anhydride;
- (7) diesters of an unsaturated dicarboxylic acid such as diethyl maleate, dibutyl maleate, di-2-ethylhexyl maleate, dibutyl fumarate and di-2-etylhexyl fumarate;
- (8)  $\alpha$ -methylene aliphatic monocarboxylic acid esters such as methyl acrylate, ethyl acrylate, n-butyl acrylate, isobutyl acrylate, dodecyl acrylate, 2-chloroethyl acrylate, phenyl acrylate, methyl  $\alpha$ -chloroacrylate, methyl methacrylate and ethyl methacrylate;
- (9) nitriles such as acrylonitrile and methacrylonitrile;
  - (10) amides such as acrylamide;
  - (11) anilides such as acrylic anilide, p-chloroacrylic

anilide, m-nitroacrylic anilide and m-methoxyacrylic anilide;

- (12) vinyl ethers such as methyl vinyl ether, ethyl vinyl ether, isobutyl vinyl ether and  $\beta$ -chloroethyl vinyl ether.
- (13) vinyl derivatives such as vinyl chloride, vinylidene chloride and vinylidene cyanide;
- (14) ethylene derivatives such as 1-methyl-1-methoxyethylene, 1,1-dimethoxyethylene, 1,2-dimethoxyethylene, 1,1-dimethoxycarbonyl ethylene and 1-methyl-1-nitroethylene;
- (15) vinyl type monomers of N-vinyl compounds such as N-vinyl pyrrole, N-vinyl carbazole, N-vinyl indole, N-vinyl pyrrolidene and N-vinyl pyrrolidene;
- (16) glycidyl acrylates such as glycidyl methyl acrylate and glycidyl methyl methacrylate; and
- (17)  $\alpha$ -cyanoacrylates such as  $\alpha$ -cyanomethyl acrylate,  $\alpha$ -cyanoethyl acrylate and  $\alpha$ -cyanomethyl methacrylate. [0166]

Among polymers prepared by these monomers as described above, preferable examples of fluorine type surfactants employed in the present invention include a copolymer of one, two or more monomers represented by any one of the formulae (1), (2) and (3), and one, two or more monomers of polyoxyalkylene (metha)acrylates, unsaturated carboxylic acids, glycidyl acrylates,  $\alpha$ -cyanoacrylates or amides. [0167]

For example, Surflon S-381, S-382, SC-101, SC-102, SC-103 and SC-104 (each manufactured by Asahi Glass Co., Ltd.), Florard FC-430, FC-431 and FC-173 (each fluorochemical, manufactured by Sumitomo 3M Ltd.), F TOP EF352, EF301 and EF303 (each manufactured by Shin Akitakasei Co., Ltd.), Schwegaufluer 8035 and 8036 (each manufactured by Schwegman Co.), BM1000 and BM1100 (each manufactured by BM Chemie), Megafac F-171 and F-177 (each manufactured by DAINIPPON INK \$ CHEMICALS, INC.) can be mentioned.

A content of each of the nonionic surfactant, amphoteric surfactant and fluorine type surfactant in the above image forming layer is preferably from 0.05 to 15% by weight and, more preferably, from 0.1 to 5% by weight. A plasticizer is further added in the image forming layer as required in order to impart the coated film with flexibility. For example, polyethylene glycol, tributyl citrate, diethyl phthalate, dibutyl phthalate, hexyl phthalate, dioctyl phthalate, tricresyl phosphate, tributyl phosphate, trioctyl phosphate and tetrahydrofurfuryl oleate are employed.

[0169]

[0168]

Silicone Type Additive

Surfactants such as silicone oil, silicone-alkyleneoxide copolymers such as L-5410 marketed from Union Carbide Company, silicone type surfactants

manufactured by Nippon Unicar Co., Ltd., silicone oil-containing aliphatic epoxides, silicon-containing monoepoxides and the like are mentioned, in order to adjust the coefficient of surface friction according to the present invention. Silicone type additives as described in any one of "New silicone and its Application" published by Toshiba Silicone in 1994, "Special Silicone Reagent Catalogue" published by AZmax Co. in 1996 and the like can be employed. An amount of any one of these additives to be added is preferably from 0.01 to 1% by weight.

[0170]

## Coating Method

As for coating methods for coating the photosensitive composition according to the present invention on a support, known methods such as spin coating, wire-bar coating, dip coating, air-knife coating, spray coating, air-spray coating, electrostatic air-spray coating, roll coating, blade coating and curtain coating are employed. An amount to be coated varies depending on applications and is preferably from  $0.05\ \text{to}\ 5.0$ g/m² as solid content. As such amount becomes smaller, apparent sensitivity becomes higher but film characteristics of the image forming layer is more deteriorated.

[0171]

It is preferable to dry a layer of the above-described photosensitive composition coated on a support with a drying

temperature of 30°C or more for 5 seconds or more and it is particularly preferable to dry it with a drying temperature of 50°C or more for 10 seconds or more. It is also preferable to treat the coated film such that remaining solvent in the surface of the image forming layer of an image forming material subjected to the above-described drying conditions become  $20 \, \text{mg/m}^2$  or less.

[0172]

# Coating Solvent

Examples of solvents to be employed for coating in the present invention are as follows:

n-propanol, isopropanol, n-butanol, sec-butanol, isobutanol, 2-methyl-1-butanol, 3-methyl-1-butanol, 2-methyl-2-butanol, 2-ethyl-1-butanol, 1-pentanol, 2-pentanol, 3-pentanol, n-hexanol, 2-hexanol, cyclohexanol, methyl cyclohexanol, 1-heptanol, 2-heptanol, 3-heptanol, 1-octanol, 4-methyl-2-pentanol, 2-hexyl alcohol, benzyl alcohol, ethylene glycol, diethylene glycol, a triethylene glycol, tetraethylene glycol, 1,3-propanediol, 1,5-pentane glycol, dimethyl triglycol, furfuryl alchol, hexylene glycol, hexyl ether, 3-methoxy-1-butanol, 3-methoxy-3-methyl butanol, butyl phenyl ether, ethylene glycol monoacetate, propylene glycol monomethyl ether, propylene glycol monomethyl ether, propylene glycol monoethyl ether, propylene glycol monopropyl ether, propylene glycol monobutyl ether, propylene glycol

phenyl ether, dipropylene glycol monomethyl ether, dipropylene glycol monoethyl ether, dipropylene glycol ether, monopropyl dipropylene glycol monobutyl ether, tripropylene glycol monomethyl ether, methyl carbitol, ethyl carbitol, ethyl carbitol acetate, butyl carbitol, triethylene glycol monomethyl ether, triethylene glycol monoethyl ether, tetraethylene glycol dimethyl ether, diacetone alcohol, acetophenone, cyclohexanone, methyl cyclohexanone, acetonyl acetone, isophorone, methyl lactate, ethyl lactate, butyl lactate, propylene carbonate, phenyl acetate, sec-butyl acetate, cyclohexyl acetate, diethyl oxalate, methyl benzoate, ethyl benzoate, γ-butyl lactone, 3-methoxy-1-butanol, 4-methoxy-1-butanol, 3-ethoxy -1-butanol, 3-methoxy-3-methyl-1-butanol, 3-methoxy-3-ethyl -1-pentanol, 4-ethoxy-1-pentanol, 5-methoxy-1-hexanol, 3-hydroxy-2-butanone, 4-hydroxy-2-butanone, 4-hydroxy-2-pentanone, 5-hydroxy-2-pentanone, 4-hydroxy-3-pentanone, 6-hydroxy-2-pentanone, 4-hydroxy-3-pentanone, 6-hydroxy-2-hexanone, 3-methyl-3-hydroxy-2-pentanone, MC, EC, allyl alcohol, isopropyl ether, butyl ether, anisole, propylene glycol monomethyl ether acetate, diethyl carbitol, tetrahydrofuran, dioxane, dioxolane, acetone, methyl propyl ketone, methyl ethyl ketone, methy amyl ketone, diethyl ketone, ethyl butyl ketone, dipropyl ketone, diisobutyl ketone,

3-hydroxy-2-butanone, 4-hydroxy-2-butanone, 2-methoxyethyl acetate, 2-ethoxyethyl acetate, methoxybutyl acetate, methyl propionate, ethyl propionate, propyl propionate, methyl butyrate, butyrate, butyl ethyl butyrate, N-methyl-2-pyrrolidone, acetonitrile, DMF, DMAc, n-pentane, 2-methyl pentane, 3-ethyl pentane, methyl cyclopentane, isohexane, cyclohexane, methyl cyclohexane, N-hexane, n-heptane, cycloheptane, n-octane, isooctane, nonane, decane, benzene, toluene, o-xylene, m-xylene, p-xylene, ethyl benzene, o-diethyl benzene, m-diethyl benzene, p-diethyl benzene, cumene, n-amyl benzene, DMDG and ethanol.

[0173]

#### Other Additives

In this cured layer, a dye, a pigment, a thickener, a plasticizer, a stabilizer, a leveling agent, a tackifier or a photobleaching inhibitor as an inactive ingredient, and a UV ray absorber as a photofading inhibitor may be contained. The Ultraviolet ray-curable protective layer mentioned above can form irregularities of fixed patterns on a surface. It is preferable to form patterns such as unevenness of a specified pattern, for example, a type of mark, a minute pattern or a background pattern on the surface of the ultraviolet ray-cured layer, since it becomes possible to identify the counterfeit, the forgery or the alteration.

[0174]

The surface of the ultraviolet ray-curable protective layer is allowed to have irregularities by a method of coating the ultraviolet ray-cured layer by using a plate having a gravure grating with the specified pattern, a method of embossing the ultraviolet ray-curing resin when it is half cured and the like.

#### [Unsaturated Group-Containing Resin]

The unsaturated group-containing resin is characterized by containing a group capable of performing polymerization by a radical or an acid. The unsaturated group denotes, for example, a glycidyl, a (metha) acryloyl or a vinyl group. The resins each containing any one of the following structures are mentioned:

(1) to (4)

[0175]

The reactive resins employed in the present invention are preferably those as represented by the above formula (1), in which  $R_1$  and  $R_2$  each represent a hydrogen atom or a methyl group. R represents a substituted or unsubstituted aryl group, a substituted or unsubstituted phenyl group or alkyl group; an alkyl group represented by  $R_3$  is preferably an alkyl group having 7 carbon atoms or less such as a methyl group or an ethyl group; an aryl group represented by  $R_3$  is preferably an aryl

group having 10 carbon atoms or less such as a phenyl group or a naphthyl group.

[0176]

As divalent linking groups represented by L,  $-CH_2-CH(OH)-CH_2-O-$ ,  $-OCH_2CH(OH)-CH_2-O-$ ,  $-OCH_2CH_2OCONH-R4-NHCOOCH_2-$  (R4 representing a p-phenylene group),  $-OCH_2CH_2OCOCH_2CH_2COOCH_2-$ ,  $-OCH_2CH_2OCO-R5-COOCH_2-$  (R5 representing an o-phenylene group) and the like are cited. [0177]

According to the present invention, L preferably represents -O-CH<sub>2</sub>CH(OH)-CH<sub>2</sub>O-CO-. The vinyl type molecular weight polymer employed in the present invention preferably contains a unit represented by the above mentioned formula (1) in an amount of from 0.001 to 20% by weight and, more preferably, from 0.001 to 10% by weight. In a case in which the content of the unit represented by the above mentioned formula (1) is less than 0.001% by weight, the cured layer is easily deteriorated in both stability against chemicals and scratch strength due to reduction of the polymerization degree, while in a case in which it is particularly more than 20% by weight, gellation is generated during synthesis, which causes a low yield as a practical problem. Or, since it has an excess amount of a reactive group, such problems as lowering the breaking elongation due to loss of flexibility of the resin itself are induced because a crosslinking density becomes

extremely high.

[0178]

Further, a content of the carboxyl group is preferably from 3 to 300 and, more preferably, from 10 to 200 in terms of an acid value. In a case in which the content of the carboxyl group is less than 3 in terms of the acid value, adhesion properties of the cured layer to a facing resin layer are deteriorated.

[0179]

A carboxy group can be introduced into the vinyl type high molecular weight polymer according to the present invention by using a method in which a monomer containing the carboxyl group such as an  $\alpha$ ,  $\beta$ -unsaturated carboxylic acid, for example, acrylic acid, methacrylic acid, maleic acid, maleic anhydride, itaconic acid, itaconic anhydride or derivatives thereof is preliminarily added as a copolymerizable component at the time of synthesizing the polymer. In a case in which employed, above mentioned derivatives are the copolymerization is once performed with an anhydride (such as maleic anhydride) and, then, the anhydride is subjected to hydrolysis by an alcohol such as methanol, ethanol, propanol or butanol, to thereby add a long-chain alkyl group to a portion of carboxylic acid on one side. Further, a method which allows an active group in the high molecular weight polymer, for example, a hydroxyl group, an amino group to perform a

polymerization reaction with a dicarboxylic acid or an acid anhydride is employed.

[0180]

The polymer containing the polymer unit represented by the formula (1) mentioned above and an  $\alpha,\beta$ -unsaturated carboxylic acid can be synthesized by synthesizing vinyl copolymer containing an  $\alpha,\beta$ -unsaturated carboxylic acid by a known method at the first step and, then, adding an unsaturated ethylenic compound containing a glycidyl group (epoxy group) at the second step. As for the unsaturated ethylenic compound containing glycidyl group (epoxy group), a glycidyl methacrylate and glycidyl acrylate can be cited representative examples. However, the polymers are not limited thereto and any unsaturated ethylenic compounds each having a glycidyl group (eposxy group) are permissible. [0181]

As for constitutional monomers contained in the vinyl type high molecular weight polymer according to the present invention other than polymer unit represented by the formula (1), monomers in the following items (1) to (17) can be mentioned:

(1) monomers each having an aromatic hydroxy group such as o-hydroxystyrene, p-hydroxystyrene, m-hydroxystyrene, o-hydroxyphenyl acrylate, p-hydroxyphenyl acrylate and m-hydroxyphenyl acrylate;

- (2) monomers each having an aliphatic hydroxy group such 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate, acrylamide, N-methylol N-methylol methacryl 4-hydroxybutyl acrylate, 4-hydroxybutyl methacrylate, methacrylate, 5-hydroxypentyl acrylate, 5-hydroxypentyl 6-hydroxyhexyl acrylate, 6-hydroxyhexyl methacrylate, N-(2-hydroxyethyl) acrylamide and N-(2-hydroxyethyl)methacrylamide hydroxyethyl and vinyl ether;
- (3) monomers each having an aminosulfonyl group such as m-aminosulfonyl phenyl methacrylate, p-aminosulfonyl phenyl methacrylate, m-aminosulfonyl phenyl acrylate, p-aminosulfonyl phenyl acrylate, p-aminosulfonyl phenyl acrylate, N-(p-aminosulfonyl phenyl) methacrylamide and N-(p-aminosulfonyl phenyl) acrylamide;
- (4) monomers each having a sulfonamide group such as  $N-(p-toluene \quad sulfonyl)$  acrylamide and  $N-(p-toluene \quad sulfonyl)$  methacrylamide;
- (5)  $\alpha,\beta$ -unsaturated carboxylic acids such as acrylic acid, methacrylic acid, maleic acid, maleic anhydride, itaconic acid and itaconic anhydride;
- (6) substituted or unsubstituted alkyl acrylates such as methyl acrylate, ethyl acrylate, propyl acrylate, butyl acrylate, amyl acrylate, hexyl acrylate, heptyl acrylate, octyl acrylate, nonyl acrylate, decyl acrylate, undecyl acrylate, dodecyl acrylate, benzyl acrylate, cyclohexyl

acrylate, 2-chloroethyl acrylate, N, N-dimethylaminoethyl acrylate and glycidyl acrylate;

- (7) substituted or unsubstituted alkyl methacrylates such as methyl methacrylate, ethyl methacrylate, propyl methacrylate, butyl methacrylate, amyl methacrylate, hexyl methacrylate, heptyl methacrylate, octyl methacrylate, nonyl methacrylate, decyl methacrylate, undecyl methacrylate, dodecyl methacrylate, benzyl methacrylate, cyclohexyl methacrylate, 2-chloroethyl methacrylate, N,N-dimethylaminoethyl methacrylate and glycidyl methacrylate;
- (8) acrylamides or methacrylamides such as acrylamide, methacrylamide, N-ethyl acrylamide, N-hexyl acrylamide, N-cyclohexyl acrylamide, N-phenyl acrylamide, N-nitrophenyl acrylamide, N-ethyl-N-phenyl acrylamide, N-(4-hydroxyphenyl) acrylamide and N-(4-hydroxyphenyl)-methacrylamide.
- (9) monomers each containing an alkyl fluoride group such as trifluoroethyl acrylate, trifluoroethyl methacrylate, tetrafluoropropyl acrylate, tetrafluoropropyl methacrylate, hexafluoropropyl methacrylate, octafluoropropyl acrylate, octafluoropropyl methacrylate, heptadecafluorodecyl acrylate, heptadecafluorodecyl methacrylate and N-butyl-N-(2-acryloxyethyl)heptadecafluorooctyl sulfonamide;
  - (10) vinyl ethers such as ethyl vinyl ether,

2-chloroethyl vinyl ether, propyl vinyl ether, butyl vinyl ether, octyl vinyl ether and phenyl vinyl ether;

- (11) vinyl esters such as vinyl acetate, vinyl chloroacetate, vinyl butylate and vinyl benzoate;
- (12) styrenes such as styrene, methyl styrene and chloromethyl styrene;
- (13) vinyl ketones such as methyl vinyl ketone, ethyl vinyl ketone, propyl vinyl ketone and phenyl vinyl ketone;
- (14) olefins such as ethylene, propylene, isobutylene, butadiene and isoprene;
- (15) N-vinylpyrrolidone, N-vinyl carbazole, 4-vinylpyridine and the like;
- (16) monomers each having a cyano group such as acrylonitrile, methacrylonitrile, 2-pentenenitrile, 2-methyl-3-butenenitrile, 2-cyanoethyl acrylate, o-cyano styrene, m-cyano styrene and p-cyano styrene; and
- (17) monomers each having an amino group such as N,N-diethylaminoethyl methacrylate, N,N-dimethylaminoethyl acrylate, N,N-dimethylaminoethyl methacrylate, polybutadiene urethane acrylate, N,N-dimethylaminopropyl acrylamide, N,N-dimethyl acrylamide, acryloyl morpholine, N-isopropyl acrylamide and N,N-diethyl acrylamide.

#### [0182]

A molecular weight of the vinyl type high molecular weight polymer according to the present invention is preferably

from 5,000 to 50,000 and, more preferably, from 7,000 to 50,000. In a case in which the molecular weight is less than 5,000, a film forming function falls down and chemical resistance tends to be deteriorated. In a case in which the molecular weight is more than 50,000, solubility of the resin to the coating solvent becomes problematic.

[0183]

Specific examples of the vinyl type high molecular weight polymers according to the present invention include following coumpounds:

(1) to (15)

The monomers or oligomers each having at least one polymerizable double bond in a molecule includes any monomers, oligomers or prepolymers each having at least one ethylenic unsaturated bond capable of performing the radical polymerization in a molecule and, on this occasion, known compounds can be employed without any particular restrictions. Specific examples thereof are as follows:

mono functional acrylates such as 2-ethylhexyl acrylate,
2-hydroxypropyl acrylate, glycerol acrylate,
tetrahydrofurfuryl acrylate, phenoxy ethylacrylate,
nonylphenoxy ethylacrylate, tetrahydrofurfuryloxy
ethylacrylate, tetrahydrofurfuryloxyhexanolide acrylate,

acrylate of ε-caprolactone adduct of 1,3-dioxane alcohol and 1,3-dioxolane acrylatė; corresponding methacrylate, itaconate, crotonate and maleate in which these acrylic acids are replaced by methacrylic acid, itaconic acid, crotonic acid maleic acid, respectively, that is, two functional acrylates of diacrylates such as ethylene glycol diacrylate, triethylene glycol diacrylate, pentaerythritol diacrylate, hydroquinone diacrylate, resorcin diacrylate, hexanediol diacrylate, neopentyl glycol diacrylate, tripropylene glycol diacrylate, diacrylate of hydroxy pivalic acid neopentyl glycol, diacrylate of neopentyl glycol adipate, diacrylate of ε-caprolactone adduct of hydroxy pivalic acid neopentyl glycol, 2-(2-hydroxy-1,1-dimethylethyl)-5-hydroxymethyl-5-ethyl-1, 3-dioxane diacrylate, tricyclodecane dimethylol acrylate, E-caprolactone adduct of tricyclodecane dimethylol acrylate and diacrylate of diglycidyl ether of 1,6-hexanediol; and corresponding methacrylate, itaconate, crotonate and maleate in which these acrylic acids are replaced by methacrylic acid, itaconic acid, crotonic acid and maleic acid, respectively, that is, polyfunctional acrylic acid esters trimethylolpropane triacrylate, ditrimethylolpropane trimethylolethane tetraacrylate, triacrylate, pentaerythritol triacrylate, pentaerythritol tetraacrylate, dipentaerythrytol tetraacrylate, dipentaerythrytol pentaacrylate, dipentaerythrytol hexaacrylate,

ε-caprolactone adduct of dipentaerythrytol hexaacrylate, pyrogallol triacrylate, propionic acid dipentaerythrytol triacrylate, propionic acid dipentaerythrytol tetraacrylate, hydroxypivalyl aldehyde-modified dimethylol propane triacrylate; and corresponding methacrylate, itaconate, crotonate and maleate in which these acrylic acids are replaced by methacrylic acid, itaconic acid, crotonic acid and maleic acid, respectively, that is, a phosphazene monomer, triethylene glycol, isocyanuric acid EO-modified diacrylate, isocyanuric acid EO-modified triacrylate, dimethylol tricyclodecane diacrylate, trimethylolpropane acrylic acid benzoate, alkylene glycol type acrylic acid modification and urethane modification acrylate. In particular, acrylate and methacrylate compounds are employed suitably among these. One or mixture of two kinds or more are employed in combination selecting from these compounds. In addition thereto, so called prepolymer, which is prepared by that acryl acid or methacrylic acid is introduced to an oligomer having suitable molecular weight and is provided with photopolymerizable property, is also employed suitable for a compound capable of addition polymerization or crosslinking. One kind prepolymer or a mixture of two or more kinds of prepolymers may be employed, or the prepolymer may be employed along with the monomer mentioned above. Examples of the prepolymer are polyester acrylates which are prepared by introducing (metha)acrylic

acid to a polyester obtained by bonding polybasic acid such as adipic acid, tri merit acid, maleic acid, phthalic acid, terephthalic acid, himic acid, malonic acid, succinic acid, glutaric acid, itaconic acid, pyromellitic acid, fumaric acid, pimelic acid, sebacic acid, dodecanoic acid and tetrahydrophthalic acid with a polyvalent alcohol such as ethylene glycol, propylene glycol, diethylene glycol, propylene oxide, 1,4-butanediol, triethylene glycol, tetraethylene glycol, polyethylene glycol, glycerin, trimethylol propane, pentaerythritol, sorbitol, 1,6-hexanediol and 1,2,6-hexanetriol; epoxy acrylates which are each prepared by introducing (meta)acrylic acid into an epoxide resin such as bisphenol A epichlorohydrin (meta) acrylic acid, phenol novolak epichlorohydrin (meta) acrylic acid; urethane acrylates which are each prepared by introducing (meta)acrylic acid into a urethane resin such as ethylene glycol adipic acid tolylene diisocyanate-2-hydroxyethyl acrylate, polyethylene glycol·tolylene diisocyanate-2-hydroxyethyl acrylate, hydroxyethyl phthalyl methacrylate xylene diisocyanate, 1,2-polybutadiene glycol·tolylene diisocyanate-2-hydroxyethyl acrylate, trimethylol propane propylene glycol tolylene diisocyanate 2-hydroxyethyl acrylate; silicone resin acrylates such as polysiloxane acrylate, polysiloxane diisocyanate.2-hydroxyethyl acrylate;

and an alkyd-modified acrylate which is prepared by introducing a (meta)acryloyl group into an oil modified alkyd resin and a spiran resin acrylate.
[0184]

Polymerizable or curable compound is compounded in the range of from 5% by weight or more (preferably, 20% by weight or more) to 97% by weight (preferably, 95% by weight or less) on the basis of the whole photosensitive layer composition. [Unsaturated Group-Containing Silicon or Fluorine Compound (Lubricant without Reducing Polymerization Degree)]

for the silicon compounds each containing unsaturated group, other than coupling agents cited herein, mentioned are vinyl-terminal polydimethylsiloxane, a vinyl-terminal diphenyl siloxane-dimethyl siloxane copolymer, vinyl methyl siloxane-dimethyl siloxane copolymer, a trimethyl siloxane group-terminal, a vinyl gum, a vinyl methyl siloxane homopolymer, a vinyl T-structure polymer, a vinyl methoxysiloxane homopolymer, an H silicone, an epoxy silicone, a methacrylate silicone, a mercapto silicone, an allyl group-modified silicone and a (meta)acryloyl-group silicone. A preferable amount of each of such silicone compounds as described above to be added is from 0.01 to 50% by weight and, more preferably, from 0.05 to 30% by weight. Specific examples thereof include MAC-2101 and MAC-2301 manufactured by Nippon Unicar Co. Ltd. Silicone additives as described in any one

of "New Silicone and its Application" published by Vinyl Toshiba Silicone in 1994 and "Special Silicone Reagent Catalogue" published by AZmax Co. in 1996 can be employed. [0185]

As for unsaturated group-containing fluorine compounds, for example, Lightester M-3F, Lightester M-4F, Lightester M-6F, Lightester FM-108 and Lightacrylate FA-108 available from Kyoei Kagaku Co., Ltd., or unsaturated fluorine compounds as mentioned above can each individually be added. A preferable amount of the silicon or fluorine compound containing an unsaturated group to be added is from 0.01 to 20% by weight and, more preferably, from 0.01 to 10% by weight.

[Method for Forming Protective Layer]

The protective layer is formed by transfer from the transfer foil according to the present invention. The surface having above mentioned image or optical variable device of the card is superposed to the transfer surface of the transfer sheet and, then, they are pressed from transfer sheet side by means of applying pressure as well as heating such as a thermal head, a heat roller or a hot stamp machine and, thereafter, the support of the transfer sheet is removed, to thereby form the protect layer.

[0186]

The protect layers or forming methods as described in any one of JP-A Nos. 08-324137, 04-247486, 04-286696, 04-320898,

05-139093, 06-072018 and the like can also be employed. [Layer Arrangement of Resin Layer]

Any resin layer or actinic ray-cured layer is permissible without any specific restrictions, so long as it is constituted with one or more layers and it contains a resin layer satisfying the breaking elongation and friction coefficient stipulated by the present invention. Particularly preferably, there is the resin layer in an outermost surface of the image recording body which has the friction coefficient within the range stipulated by the present invention. Thickness of the resin layer is preferably from 3 to 30  $\mu m$  and, more preferably, 3 to 5  $\mu m$ .

[0187]

[Advantage of the Invention]

As has been described above, according to the image recording body and the production method for the image recording body of the present invention, it is possible not only to enhance security against the forgery, the alteration and the like, but also to prevent scratches or wrinkles from being generated and, further, it is possible to prevent blocking to be caused during a storage in an extended period of time and, still further, to prevent the forgery or the alteration without impairing printability of characters and durability and, even still further, to easily fabricate it into a card form.

[Brief Description of Drawings]

- [FIG. 1] It is a diagram showing a layer arrangement of an image recording body.
- [FIG. 2] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
  - [FIG. 3] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 4] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 5] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 6] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 7] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 8] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 9] It is a diagram showing a layer arrangement of another embodiment of the image recording body.
- [FIG. 10] It is a diagram showing a layout of an image recording body.
- [FIG. 11] It is a diagram showing a layout of another embodiment of the image recording body.
- [FIG. 12] It is a diagram showing a layout of another embodiment of the image recording body.

- [FIG. 13] It is a diagram showing a layout of another embodiment of the image recording body.
- [FIG. 14] It is a diagram showing an embodiment of a transparent protective transfer foil.
- [FIG. 15] It is a diagram showing an embodiment of an optically variable device transfer foil.
- [FIG. 16] It is a diagram showing an embodiment of a resin transfer foil.
- [FIG. 17] It is a diagram showing an embodiment of a curing-type resin layer-containing optically variable device transfer foil.
- [FIG. 18] It is a diagram showing an arrangement of another embodiment of the image recording body.

[Description of Reference Numerals and Signs]

- 1 substrate
- 2 information-carrying layer
- 3 bead-containing layer
- 4 actinic ray-cured layer
- 5 reflection layer
- 7 printed layer

Fig. 1

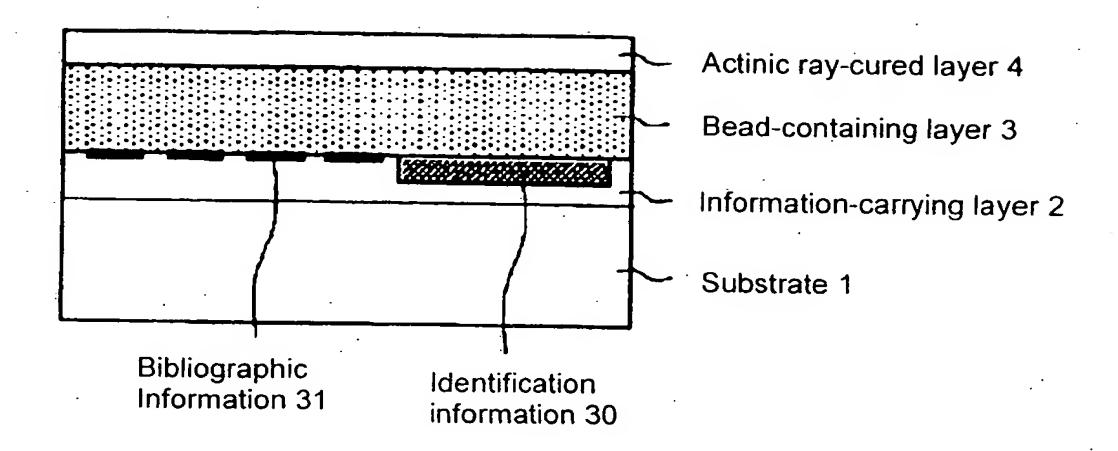


Fig. 2

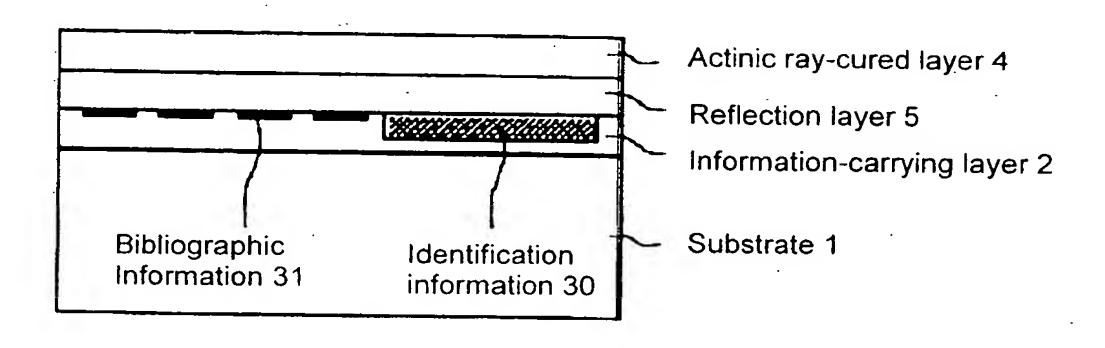


Fig. 3

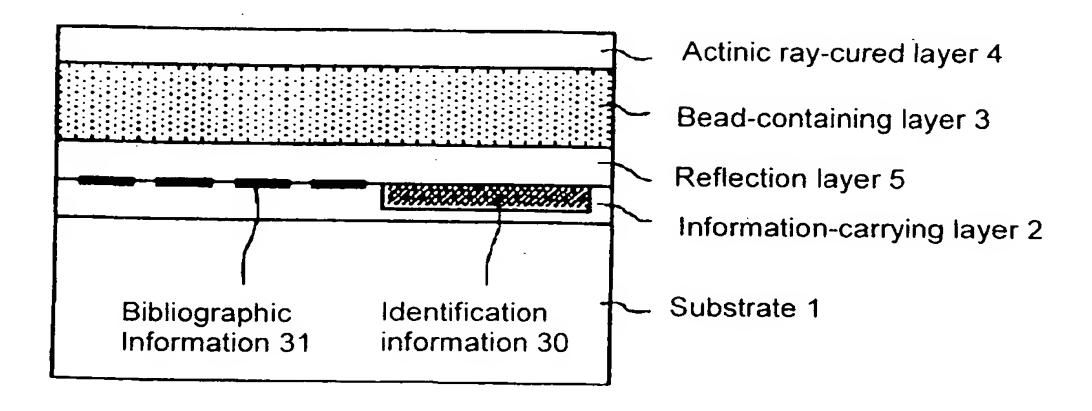


Fig. 4

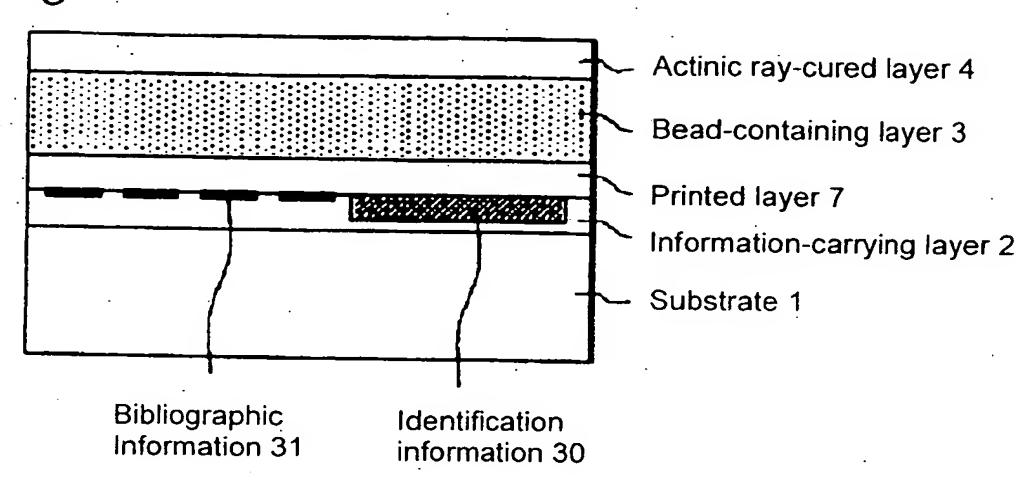


Fig. 5

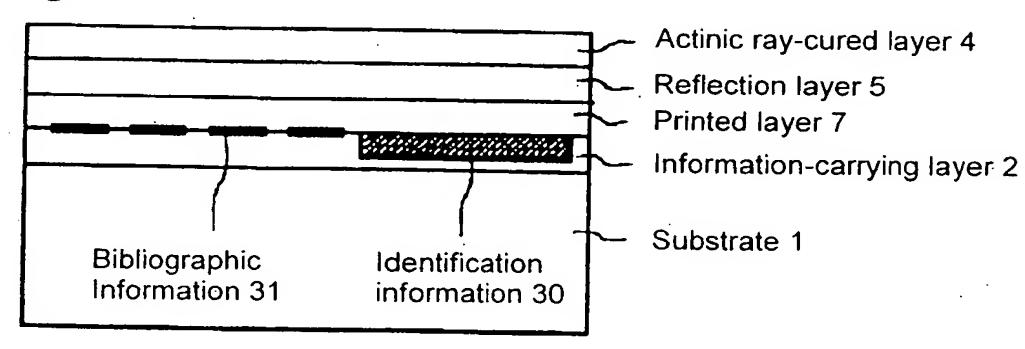


Fig. 6

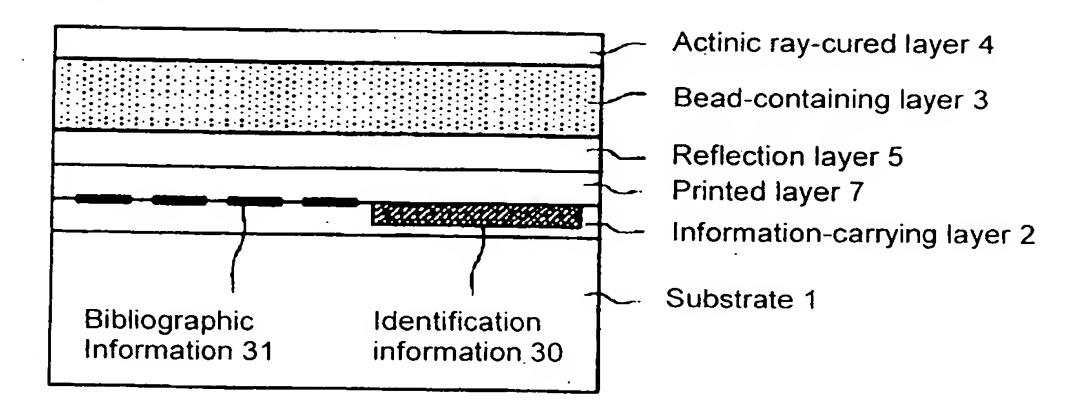


Fig. 7

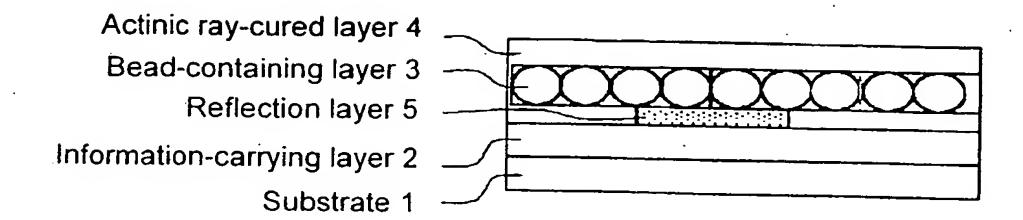


Fig. 8

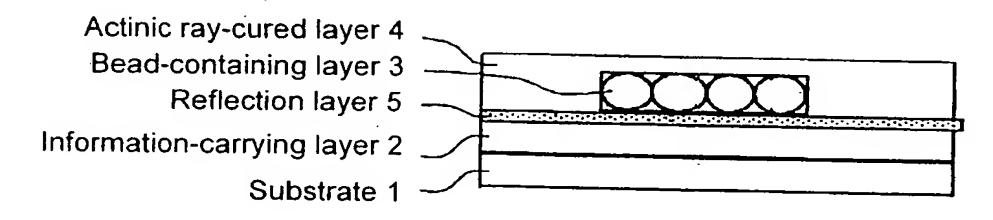
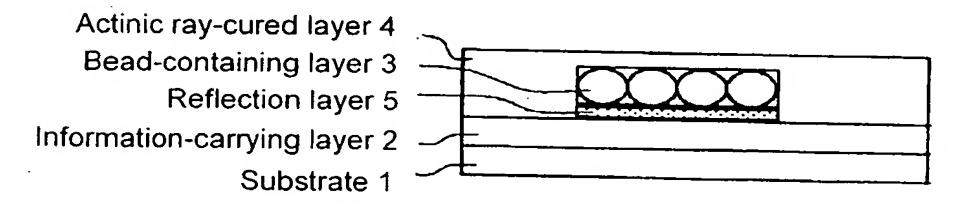


Fig. 9



Bibliographic Information 31

Bead-containing layer 3

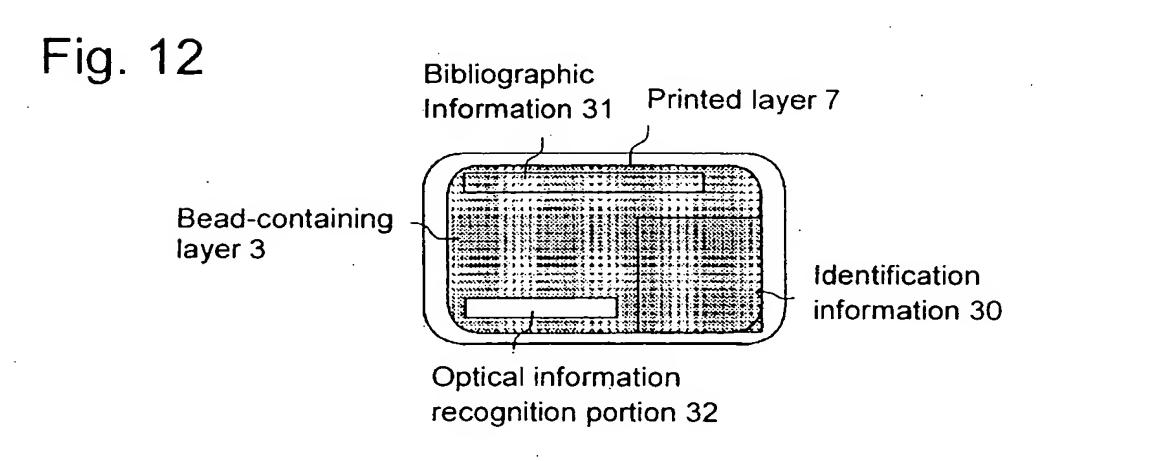
Optical information recognition portion 32

Fig. 11

Bibliographic Information 31

Identification

information 30



Optical information

recognition portion 32

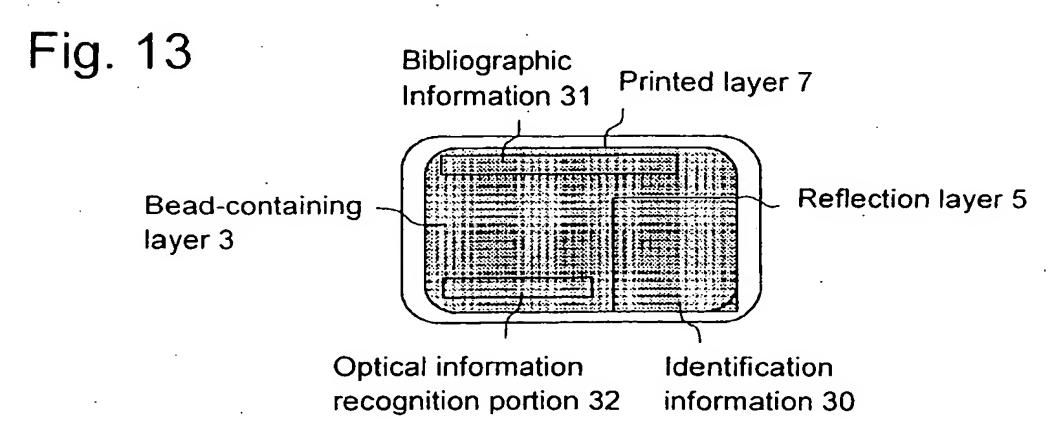


Fig. 16

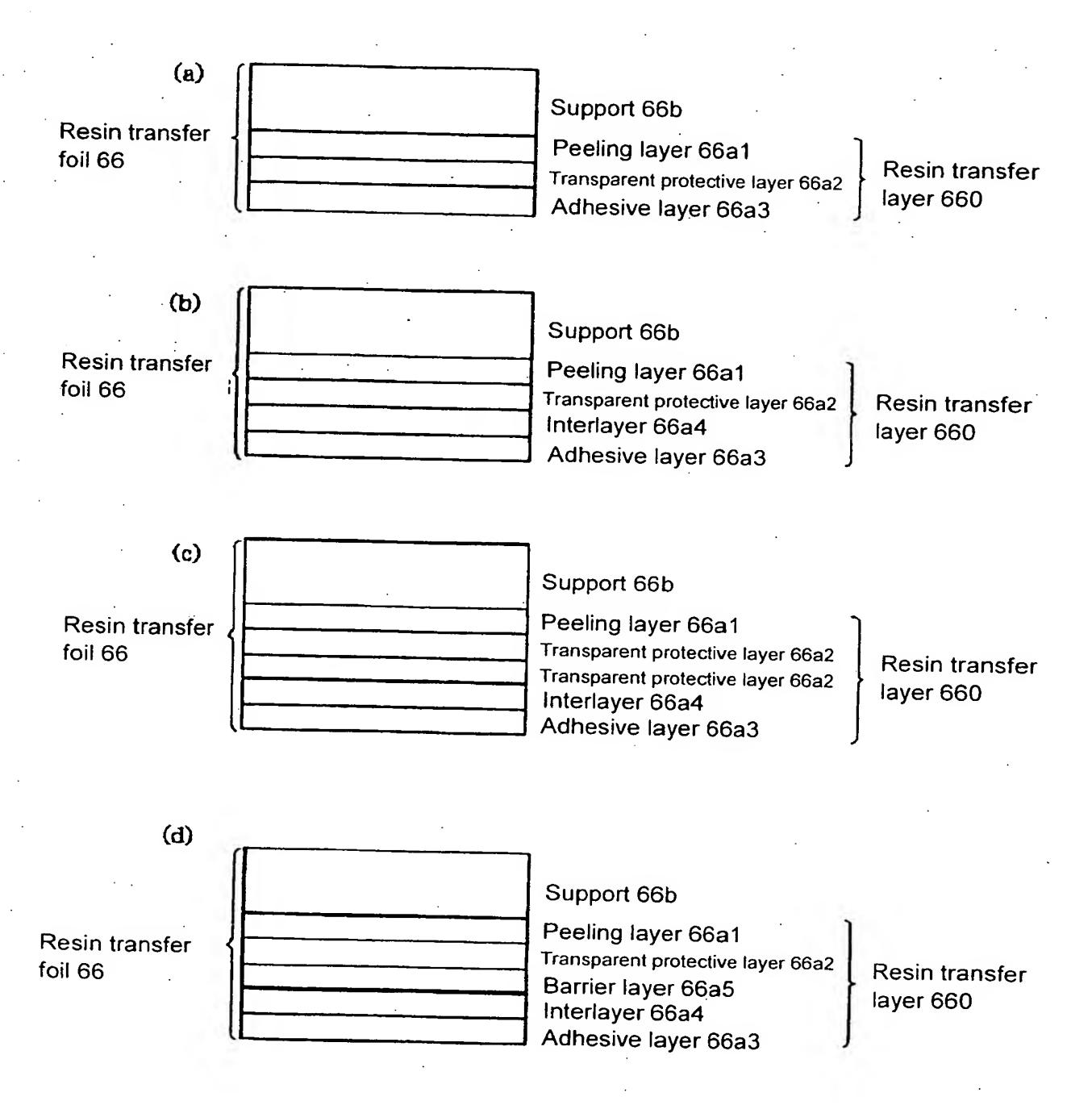
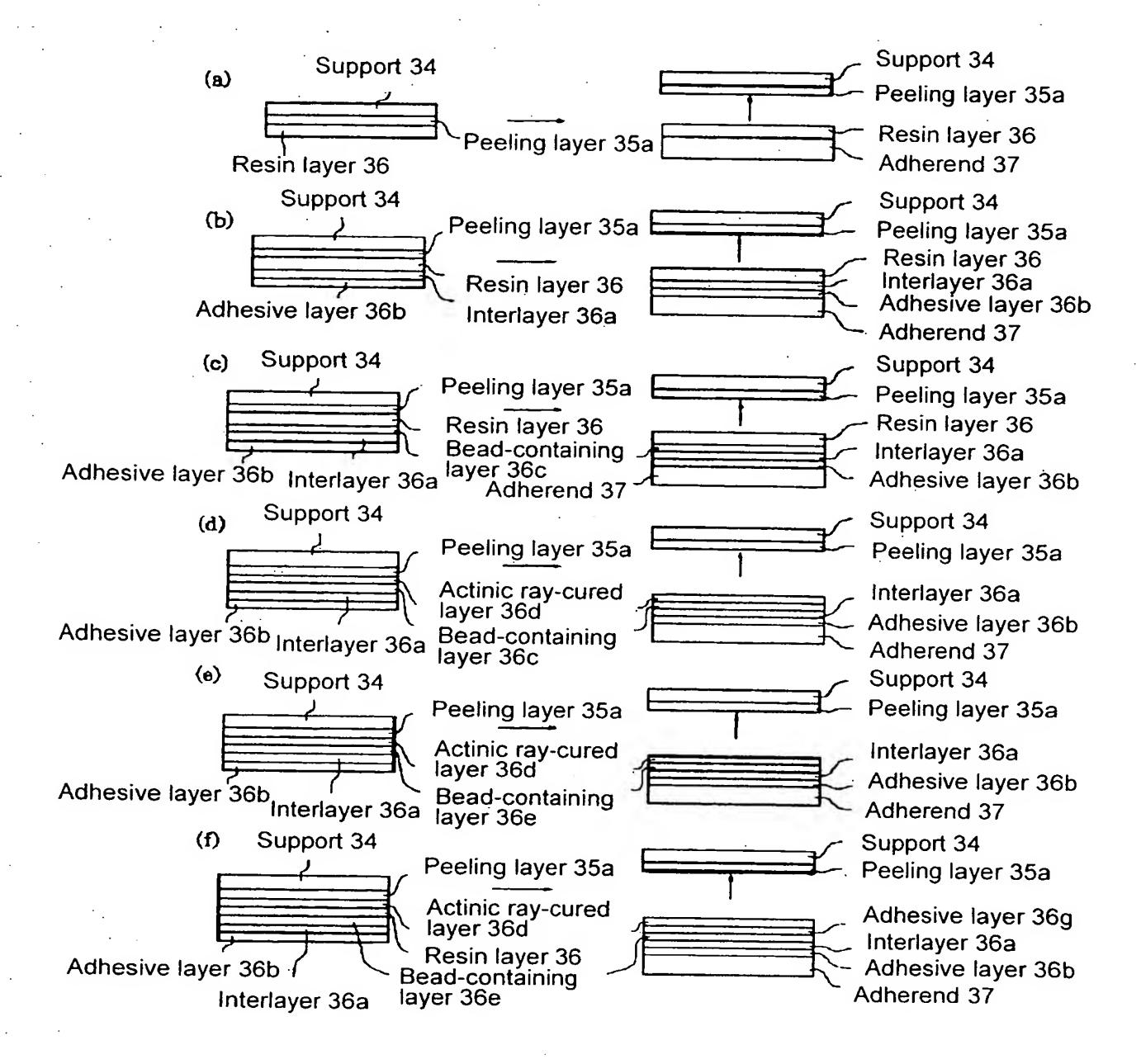


Fig. 18



#### Abstract of JP2001215304

PROBLEM TO BE SOLVED: To provide optical parts having excellent optical characteristics and having an antireflection layer excellent in durability. SOLUTION: Optical parts having excellent optical characteristics and having an antireflection film excellent in durability can be obtained. Because the optical parts have an antireflection film having high transmittance for rays and excellent antireflection performance and durability, they can be preferably used for various use.

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#### (54) 【発明の名称】 光学部品

#### (57)【要約】

(21)出願番号

【解決手段】 本発明の光学部品は、特定の環状炭化水素系重合体からなる成形体の表面の少なくとも一部に、反射防止膜が形成されてなることを特徴としている。

【効果】 本発明によれば、光学特性に優れ、かつ、耐久性に優れた反射防止膜を有する光学部品を提供することができる。このような光学部品は、光線透過率が高く、反射防止性能に優れ、かつ耐久性に優れた反射防止膜を有するため、種々の光学部品用途に好適に使用することができる。



#### 【特許請求の範囲】

【請求項1】下記(I)、(II)、(III)、(IV)およ び(V)よりなる群から選ばれる少なくとも1種の環状 炭化水素系重合体からなる成形体の表面の少なくとも一 部に、反射防止膜が形成されてなることを特徴とする光 学部品;

(I) ビニル系単環脂環族炭化水素化合物の(共) 重合 体、または、

ビニル系単環脂環族炭化水素化合物と、これと共重合可 能な単量体との共重合体、

あるいはこれらの水素添加物:

(II) ビニル系芳香族炭化水素化合物の(共) 重合体の 水素添加物、または、

ビニル系芳香族炭化水素化合物と、これと共重合可能な 単量体との共重合体の水素添加物であって、芳香族炭化 水素環の少なくとも30%が水素添加されてなる重合 体;

(III) 単環環状モノオレフィン系化合物の(共) 重合 体、または、単環環状モノオレフィン系化合物と、これ と共重合可能な単量体との共重合体;

(IV) 単環環状共役ジエン系化合物の(共) 重合体の水 素添加物、または、単環環状共役ジエン系化合物と、こ れと共重合可能な単量体との共重合体の水素添加物;

(V) 上記(I)、(II)、(III) および(IV) よりな る群から選ばれる少なくとも1種の環状炭化水素系重合 体を、 $\alpha$ ,  $\beta$  一不飽和カルボン酸、その無水物またはそ の誘導体でグラフト変性して得られる変性物。

【請求項2】反射防止膜が、屈折率の異なる複数の層か らなる積層膜である、請求項1に記載の光学部品。

【請求項3】反射防止膜が、

金属酸化物、金属硫化物または金属弗化物からなる高屈 折率層と、

金属酸化物、金属弗化物または酸化ケイ素からなる低屈・ 折率層とからなる積層膜である、請求項1に記載の光学 部品。

【請求項4】反射防止膜が、

酸化ケイ素からなる層と、

金属酸化物、金属硫化物または金属弗化物からなる高屈 折率層と、

折率層とからなる積層膜である、請求項1に記載の光学 部品。

#### 【発明の詳細な説明】

[0001]

【産業上の利用分野】本発明は光学部品に関し、詳しく は、耐久性に優れた反射防止層を有する工学部品に関す る。

[0002]

【発明の技術的背景】従来、各種の透明プラスチック材 料が、光学用レンズ、メガネレンズ、光ディスク用基

板、プラスチック光ファイバなどの光学部品用原料とし て用いられている。

【0003】たとえば、透明プラスチック材料から、射 出成形などの方法で非球面レンズが製造されており、レ ーザー光用、カメラ用、スクリーン投影用などの各種用 途で使用されている。

【0004】このような射出成形によるレンズ用の樹脂 としては、ポリメチルメタクリレート (PMMA)、ポ リアクリレート系共重合体、ポリカーボネート、ノルボ 10 ルネン系樹脂などが知られている。

【0005】しかしながら、ポリメチルメタクリレート やポリアクリレート系共重合体は、吸水性が比較的大き いため、レンズなどの光学部品として用いた場合には、 吸湿による光学特性の変化を生じやすく、精密な用途に は用いられないという問題がある。また、ポリカーボネ ートは複屈折が大きく、表面硬度が十分でないという問 題がある。

【0006】一方、レンズなどの光を透過させる光学部 品の表面には、誘電体多層膜である反射防止膜を形成し て使用することが知られている。これは一般に、基材を 成形しただけのレンズでは、基材の屈折率と光の入射媒 質である空気との屈折率の差に起因して、基材表面で入 射光が一部反射し、さらに、基材からの出射界面におい ても反射するため、反射損失を生じてしまうという問題 があるが、レンズ表面に反射防止膜を設けることによ り、反射損失を低減させることができるためである。そ して、プラスチックレンズの表面に形成する反射防止膜 としては、金属酸化物、金属弗化物などの蒸着膜が知ら れている。

【0007】しかしながら、上述した樹脂から形成され たプラスチックレンズの表面に、金属酸化物などによる 反射防止膜を形成した場合には、反射防止膜の密着性が 充分ではなく、基材からの剥離を生じやすく、耐久性が 悪いという問題がある。

【0008】このため、優れた光学特性を示し、かつ、 耐久性に優れた反射防止膜を有する光学部品の出現が強 く求められていた。本発明者は、このような状況に鑑み て鋭意研究したところ、特定の環状炭化水素系重合体か らなる成形体が、透明性に優れ複屈折が小さく、光学特 金属酸化物、金属弗化物または酸化ケイ素からなる低屈 40 性に優れており、かつ、反射防止膜との密着性に優れる ことを見出して本発明を完成するに至った。

[0009]

【発明の目的】本発明は、優れた光学特性を有し、かつ 耐久性に優れた反射防止層を有する光学部品を提供する ことを目的としている。

[0010]

【発明の概要】本発明の光学部品は、下記(I)、(I I)、(III)、(IV)および(V)よりなる群から選ば れる少なくとも 1 種の環状炭化水素系重合体からなる成 50 形体の表面の少なくとも一部に、反射防止膜が形成され



てなることを特徴としている。

- (I) ビニル系単環脂環族炭化水素化合物の(共) 重合 体、または、ビニル系単環脂環族炭化水素化合物と、こ れと共重合可能な単量体との共重合体、あるいはこれら の水素添加物;
- (II) ビニル系芳香族炭化水素化合物の(共) 重合体の 水素添加物、または、ビニル系芳香族炭化水素化合物 と、これと共重合可能な単量体との共重合体の水素添加 物であって、芳香族炭化水素環の少なくとも30%が水 素添加されてなる重合体;
- (III) 単環環状モノオレフィン系化合物の(共) 重合 体、または、単環環状モノオレフィン系化合物と、これ と共重合可能な単量体との共重合体;
- (IV) 単環環状共役ジェン系化合物の(共) 重合体の水 素添加物、または、単環環状共役ジエン系化合物と、こ れと共重合可能な単量体との共重合体の水素添加物;
- (V) 上記(I)、(II)、(III) および(IV) よりな る群から選ばれる少なくとも 1 種の環状炭化水素系重合 体を、 $\alpha$ ,  $\beta$  - 不飽和カルボン酸、その無水物またはそ の誘導体でグラフト変性して得られる変性物。
- 【0011】また、本発明の好ましい態様においては、 前記反射防止膜が、屈折率の異なる複数の層からなる積 層膜であるのが好ましく、特に、前記反射防止膜が、金 属酸化物、金属硫化物または金属弗化物からなる高屈折 率層と、金属酸化物または金属弗化物からなる低屈折率 層とからなる積層膜であるか、または、酸化ケイ素から なる層と、金属酸化物、金属硫化物または金属弗化物か らなる高屈折率層と、金属酸化物または金属弗化物から なる低屈折率層とからなる積層膜であるのが望ましい。 [0012]

【発明の具体的説明】以下、本発明について具体的に説 明する。

#### 環状炭化水素系重合体

本発明で用いられる環状炭化水素系重合体は、下記 (I) 、 (II) 、 (III) 、 (IV) および (V) よりなる 群から選ばれる少なくとも 1 種の環状炭化水素系重合体 を含有する樹脂である。

- (I)ビニル系単環脂環族炭化水素化合物の(共)重合 体、または、ビニル系単環脂環族炭化水素化合物と、こ の水素添加物;
- (II)ビニル系芳香族炭化水素化合物の(共)重合体の 水素添加物、または、ビニル系芳香族炭化水素化合物 と、これと共重合可能な単量体との共重合体の水素添加 物であって、芳香族炭化水素環の少なくとも30%が水 素添加されてなる重合体;
- (III)単環環状モノオレフィン系化合物の(共)重合 体、または、単環環状モノオレフィン系化合物と、これ と共重合可能な単量体との共重合体;
- (IV) 単環環状共役ジエン系化合物の(共) 重合体の水 50

素添加物、または、単環環状共役ジエン系化合物と、こ れと共重合可能な単量体との共重合体の水素添加物;

(V) 上記(I)、(II)、(III) および(IV) よりな る群から選ばれる少なくとも1種の環状炭化水素系重合 体を、 $\alpha$ ,  $\beta$  - 不飽和カルボン酸、その無水物またはそ の誘導体でグラフト変性して得られる変性物。

【0013】ここで、本発明で用いられる環状炭化水素 系重合体として用いることのできる、上記(I)、(I I) 、 (III) 、 (IV) および (V) の重合体について説 明する。

【0014】(I)の重合体は、(I)ビニル系単環脂環 族炭化水素化合物の(共)重合体、または、ビニル系単 環脂環族炭化水素化合物と、これと共重合可能な単量体 との共重合体、あるいはこれらの水素添加物である。

【0015】ビニル系単環脂環族炭化水素化合物とは、 ビニル基または、αーアルキル置換ビニル基に、単環の シクロアルキル基または単環のシクロアルケニル基が結 合した構造を有する化合物である。ここで該単環のシク ロアルキル基およびシクロアルケニル基は、アルキル基 20 などの置換基を有していてもよい。

【0016】このような化合物としては、ビニルシクロ ブタン、ビニルシクロペンタン、ビニルシクロヘキサ ン、ビニルシクロヘプタン、ビニルシクロオクタンおよ びこれら化合物のビニル基のα位がメチル、エチル、プ ロピルなどのアルキル基で置換された化合物が例示でき る。また、4ービニルシクロヘキセン、4ーイソプロペ ニルヘキセン、1-メチルー4-ビニルシクロヘキセ ン、1ーメチルー4ーイソプロペニルシクロヘキセン、 2ーメチルー4ービニルシクロヘキセン、2ーメチルー 30 4ーイソプロペニルヘキセンなどのビニルシクロヘキセ ン誘導体を例示できる。

【0017】上記(I)の重合体が、ビニル系単環脂環 族炭化水素化合物の(共)重合体である場合には、これ らのビニル系単環脂環族炭化水素化合物が単独で重合し た重合体であっても、2種以上組み合わせて共重合した 共重合体であってもよい。.

【0018】また、上記(I)の重合体が、ビニル系単 環脂環族炭化水素化合物と、これと共重合可能な単量体 との共重合体である場合には、これらのビニル系単環脂 れと共重合可能な単量体との共重合体、あるいはこれら 40 環族炭化水素化合物と、その化合物と共重合可能な他の 化合物とが、本発明の趣旨を損なわない範囲で組み合わ せて共重合された共重合体である。

> 【0019】ビニル系単環脂環族炭化水素化合物と共重 合する単量体としては、共重合可能な単量体をいずれも 用いることができ、プロピレン、プテン、アクリロニト リル、アクリル酸、メタクリル酸、無水マレイン酸、ア クリル酸エステル、メタクリル酸エステル、マレイミ・ ド、酢酸ビニル、塩化ビニルなどを例示することができ る。このような単量体の中でも、αーオレフィンを用い るのが好ましく、特に、プロピレン、ブテンのような単



量体を用いると、柔軟性や耐衝撃性を付与できるので好ましい。このような、ビニル系単環脂環族炭化水素化合物と共重合可能な単量体は、単量体総量に対して0~95モル%、より好ましくは0~90モル%となる割合で用いられるのが望ましい。

【0020】これらの重合体あるいは共重合体を得るための重合方法に特に制限はなく、公知のラジカル重合、配位アニオン重合(チーグラー重合)、カチオン重合、アニオン重合などの重合方法が適用できる。

【0021】また、上記(I)の重合体は、これらの重合体あるいは共重合体の水素添加物であってもよい。特に、これらの重合体あるいは共重合体を形成する単量体として用いられるビニル系単環脂環族炭化水素化合物が、ビニル基またはαーアルキル置換ビニル基に、単環の(アルキル置換)シクロアルケニル基が結合した構造を有する化合物である場合には、上述の重合体あるいは共重合体は水素添加物であるのが好ましい。このような水素添加物としては、重合体あるいは共重合体中の置換基を有していてもよいシクロアルケニル基の30%以上、好ましくは60%以上、さらに好ましくは90%以上が水素添加されてなる重合体が望ましい。

【0022】(II)の重合体は、ビニル系芳香族炭化水素化合物の(共)重合体の水素添加物、または、ビニル系芳香族炭化水素化合物と、これと共重合可能な単量体との共重合体の水素添加物であって、芳香族炭化水素環の少なくとも30%が水素添加されてなる重合体である。

【0023】(II)の重合体を形成する単量体として用いられる、ビニル系芳香族炭化水素化合物は、ビニル基または $\alpha$ -アルキル置換ビニル基に、芳香族系置換基が 30結合した化合物である。このような化合物としては、スチレン、 $\alpha$ -メチルスチレン、 $\alpha$ -イソプロピルスチレン、 $\alpha$ -インプロピルスチレン、 $\alpha$ -インプロピルスチレン、 $\alpha$ -オーブチルスチレン、 $\alpha$ -インプロピルスチレン、 $\alpha$ -オーブチルスチレン、 $\alpha$ -オーブチルスチレン、 $\alpha$ -ガーン、 $\alpha$ 

【0024】上記(II)の重合体が、ビニル系芳香族炭化水素化合物の(共)重合体の水素添加物である場合には、このようなビニル系芳香族炭化水素化合物が単独で重合した重合体の水素添加物であっても、2種以上組み合わせて共重合した共重合体の水素添加物であってもよい。

【0025】また、上記(II)の重合体が、ビニル系芳香族炭化水素化合物と、これと共重合可能な単量体との共重合体の水素添加物である場合には、このようなビニル系芳香族炭化水素化合物と、その化合物と共重合可能 50

な単量体とが、本発明の趣旨を損なわない範囲で組み合わせて共重合された共重合体の水素添加物である。このようなビニル系芳香族炭化水素化合物と共重合する単量体としては、共重合可能な単量体をいずれも用いることができ、プロピレン、ブテン、アクリロニトリル、アクリル酸、メタクリル酸、無水マレイン酸、アクリル酸エステル、メタクリル酸エステル、マレイミド、酢酸ビニル、塩化ビニルなどを例示することができる。このような単量体の中でも、αーオレフィンを用いるのが好ましく、行ロピレン、ブテンのような単量体を用いると、柔軟性や耐衝撃性を付与できるので好ましい。このような、ビニル系芳香族炭化水素化合物と共重合可能な単量体は、単量体総量に対して0~95モル%、より好ましくは0~90モル%となる割合で用いられるのが望ましい。

【0026】このような、上記(II)の重合体あるいは 共重合体は、上記(I)の重合体と同様の方法で重合 し、公知の方法で水素添加することによって得ることが できる。

【0027】水素添加の方法としては、たとえば、特開平7-247321、米国特許5,612,422等の方法などを例示できる。上記(II)の重合体中に含まれる芳香族炭化水素環の水素添加率は、30%以上、好ましくは60%以上、より好ましくは90%以上である。【0028】(III)の重合体は、単環環状モノオレフィン系化合物の(共)重合体、または、単環環状モノオレフィン系化合物と、これと共重合可能な単量体との共重合体である。

【0029】(III) の重合体を形成する単量体として 用いられる、単環環状モノオレフィン系化合物は、置換 基を有していてもよい単環の環状モノオレフィンであっ て、シクロブテン、シクロペンテン、シクロヘキセン、 シクロオクテンなどを挙げることができる。

【0030】上記(III)の重合体が、単環環状モノオレフィン系化合物の(共)重合体である場合には、このような単環環状モノオレフィン系化合物が単独で重合した重合体であっても、2種以上組み合わせて共重合した重合体であってもよい。

【0031】また、上記(III)の重合体が、単環環状モノオレフィン系化合物と、これと共重合可能な単量体との共重合体である場合には、上記した単環環状モノオレフィン系化合物と、その化合物と共重合可能なほかの化合物とが、本発明の趣旨を損なわない範囲で組み合わせて共重合された共重合体である。

【0032】単環環状モノオレフィン系化合物と共重合する単量体としては、共重合可能な単量体をいずれも用いることができるが、エチレン、プロピレン、ブテン、アクリロニトリル、アクリル酸、メタアクリル酸、無水マレイン酸、アクリル酸エステル、メタクリル酸エステル、マレイミド、酢酸ビニル、塩化ビニル等が例示でき





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る。このような、単環環状モノオレフィン系化合物と共 重合可能な単量体は、単量体総量に対して0~95モル %、より好ましくは0~90モル%となる割合で用いら れるのが望ましい。

【0033】このような、上記(III)の重合体あるいは共重合体を得るための重合方法に特に制限はなく、環状モノオレフィン系化合物を含む単量体を付加重合する、公知の方法によって得ることができる。

【0034】(IV)の重合体は、単環環状共役ジェン系化合物の(共)重合体の水素添加物、または、単環環状 10 共役ジェン系化合物と、これと共重合可能な単量体との共重合体の水素添加物である。

【0035】(IV)の重合体を形成する単量体として用いられる、単環環状共役ジェン系化合物は、置換基を有していてもよい単環環状共役ジェンであって、シクロペンタジェン、シクロヘキサジェン、シクロヘプタジェン、シクロオクタジェンなどを挙げることができる。

【0036】上記(IV)の重合体が、単環環状共役ジエン系化合物の(共)重合体の水素添加物である場合には、このような単環環状共役ジエン系化合物を単独で重 20合した重合体の水素添加物であっても、2種以上組み合わせて共重合した共重合体の水素添加物であってもよい。

【0037】また、上記(IV)の重合体が、単環環状共役ジエン系化合物と、これと共重合可能な単量体との共重合体の水素添加物である場合には、このような単環環状共役ジエン系化合物と、その化合物と共重合可能な単量体とが、本発明の趣旨を損なわない範囲で組み合わせて共重合された共重合体の水素添加物である。

【0038】このような単環環状共役ジエン系化合物と 30 共重合する単量体としては、共重合可能な単量体をいず れも用いることができるが、エチレン、プロピレン、ブ テン、アクリロニトリル、アクリル酸、メタアクリル 酸、無水マレイン酸、アクリル酸エステル、メタクリル 酸エステル、マレイミド、酢酸ビニル、塩化ビニル等が 例示できる。このような単環環状共役ジエン系化合物と 共重合可能な単量体は、単量体総量に対して0~95モ ル%、より好ましくは0~90モル%となる割合で用い られるのが望ましい。

【0039】このような、上記(IV)の重合体あるいは 40 共重合体は、上述した単環環状共役ジエン系化合物を含む単量体を公知の方法で付加重合し、公知の方法で水素 添加することによって得ることができる。

【0040】具体的には、たとえば、ポリシクロヘキサジエンとそれからなる水素添加物は特開平11-106571で開示されている方法を用いて入手することができる。このような、上記(IV)の重合体あるいは共重合体中に含まれる、炭化水素環中の二重結合の水素添加率は、30%以上、好ましくは60%以上、より好ましくは90%以上であるのが望ましい。

【0041】 このような、(I)、(II)、(III) およ び(IV)の環状炭化水素系重合体は、ゲルパーミエーシ ョンクロマトグラフィー(GPC)による重量平均分子 量(Mw)が、ポリスチレン換算で通常5,000~ 1,000,000、好ましくは10,000~50 0,000、より好ましくは50,000~300,0 .00であるのが望ましい。また、分子量分布Mw/Mn は、10以下、好ましくは5.0以下、より好ましくは 3. 0以下であるのが望ましく、また、ガラス転移温度 は、50~300°C、好ましくは60~280°C、より 好ましくは、70~250℃の範囲にあり、結晶化度 は、20%以下、好ましくは10%以下、より好ましく は5%以下であるのが望ましい。また、密度は、1.5 g/cm³以下、好ましくは1.0g/cm³以下、より 好ましくは O. 9 5 g/c m³以下であるのが望まし い。

【0042】(V)の重合体は、上記(I)、(II)、(III)、(III) および(IV)よりなる群から選ばれる少なくとも 1種の環状炭化水素系重合体を、 $\alpha$ ,  $\beta$  —不飽和カルボン酸、その無水物またはその誘導体でグラフト変性して得られる変性物である。

【0043】(V)の重合体は、このような変性前の環 状炭化水素系重合体を、グラフトモノマーを用いてグラ フト変成したものである。グラフトモノマーとしては、 たとえば、アクリル酸、メタクリル酸、マレイン酸、フ マル酸、テトラヒドロフタル酸、イタコン酸、シトラコ ン酸、クロトン酸、ナジック酸などの $\alpha$ ,  $\beta$  一不飽和カ ルボン酸、またはこれらの酸無水物あるいはこれらの誘 導体を用いることができ、誘導体としては、たとえば、 酸ハライド、アミド、イミド、エステルなどが挙げられ る。このような無水物あるいは誘導体としては、具体的 には、塩化マレニル、マレニルイミド、無水マレイン 酸、無水シトラコン酸、マレイン酸モノメチル、マレイ ン酸ジメチルなどが挙げられる。これらの中では、不飽 和ジカルボン酸またはその酸無水物が好ましく、特にマ レイン酸、ナジック酸またはこれらの酸無水物が好適で ある。

【0044】(V)の重合体を調製する場合におけるグラフトモノマーのグラフト割合は、(I)~(IV)から選ばれる環状炭化水素系重合体1gに対して、0.001~0.04mg当量、好ましくは0.005~0.03mg当量の範囲であるのが望ましい。

【0045】また、グラフトモノマーとして、上記のような $\alpha$ ,  $\beta$  一不飽和カルボン酸などの代わりに、エポキシ変性で用いられる変性剤、たとえばグリシジルメタクリレート (GMA)、アリルグリシジルエーテル、ビニルグリシジルエーテル、グリシジルイタコネートなどのグリシジル化合物を用いることもできる。

【0046】このようなグラフトモノマーを用いて、(V)の重合体を製造するには、従来公知の種々の方法



を用いることができる。このような方法としては、たと えば、上述した(I)~(IV)から選ばれる環状炭化水 素系重合体を加熱溶融させ、グラフトモノマーを添加し てグラフト共重合する方法、あるいは、上述した(I) ~(IV)から選ばれる環状炭化水素系重合体を溶媒に溶 解させ、グラフトモノマーを添加してグラフト共重合す る方法が挙げられる。

【0047】グラフト共重合反応は、ラジカル開始剤の 存在下で行うと、反応効率がよく好ましい。グラフト共 重合反応に用いるラジカル開始剤としては、有機ペルオ キシド、有機ペルエステル、その他アゾ化合物が挙げら れる。これらラジカル開始剤の中でもジクミルペルオキ シド、ジーtertーブチルペルオキシド、2,5ージメチル -2,5-ジ(tert-ブチルペルオキシ) ヘキシン-3、 2,5-ジメチルー2,5-ジ (tert-ブチルペルオキシ) へ ・キサン、1,4ービス(tertーブチルペルオキシイソプロ ピル)ベンゼンなどのジアルキルペルオキシドが好まし い。このようなラジカル開始剤は、上述した(I)~(I V) から選ばれる環状炭化水素系重合体 100重量部に 対して、0.001~1重量部の割合で用いるのが望ま しい。

【0048】このようなグラフト共重合反応は、通常6 0~380℃の温度範囲で行われる。このような(V) の重合体は、重量平均分子量が、10,000~50 0,000であり、好ましくは20,000~300. 000、より好ましくは50,000~300,000 程度であるのが望ましい。

【0049】本発明では、このような環状炭化水素系重 合体を用いることによって、特に、光学特性に優れ、反 射防止層との密着性がよく優れた耐久性を有する光学部 30 品を得ることができる。

【0050】本発明において、成形体を形成する樹脂 は、上記のような環状炭化水素系重合体のみであること が好ましいが、環状炭化水素系重合体の特性を損なわな い範囲で、他の樹脂が配合されていてもよい。成形体を 形成する樹脂に配合することのできる他の樹脂として は、ポリエチレン、ポリプロピレン、ポリ1‐ブテン、 ポリ4‐メチル‐1‐ペンテンなどのポリオレフィン;ポ リスチレンなどを挙げることができる。

【0051】また、本発明において、成形体を形成する 樹脂には、必要に応じて老化防止剤、紫外線安定剤、帯 電防止剤、滑剤などの各種添加剤を、目的を損なわない 範囲で添加してもよい。

【0052】このような環状炭化水素系重合体からなる 成形体は、どのような方法で成形して得てもよく、たと えば射出成形、溶融押し出し、熱プレス、溶剤キャス ト、インフレーションなどの公知の方法により成形する ことができる。

#### 【0053】 反射防止膜

らなる成形体の表面の少なくとも一部に、反射防止膜が 形成されてなるものである。

【0054】本発明の光学部品における反射防止膜は、 用途により透過すべき波長の光を透過し、光学部品に入 射または出射する光の反射を防止して、光学部品の光の 反射損失を低減させることのできる膜であるのが望まし

【0055】本発明における反射防止膜は、このような 機能を有するものであれば特に限定されるものではない が、目的とする波長の光を80%以上、好ましくは90・ %以上、特に好ましくは95%以上透過するのが望まし いる

【0056】本発明における反射防止膜は、単層で形成 されていてもよいが、屈折率の異なる複数の層からなる 積層膜であると、反射防止効果が高いため好ましい。本 発明では、反射防止膜が、屈折率の異なる複数の層から なる積層膜である場合には、積層の順序を特に規定する ものではなく、2層からなる膜であっても、3層以上の 多層からなる膜であってもよい。このような反射防止膜 としては、成形体側に高屈折率の層、外側に低屈折率の 層を有する積層膜であるのが好ましい。

【0057】また本発明では、反射防止膜が、高屈折率 の層を成形体と接する層(第1層)とし、ついで低屈折 率の層(第2層)を有する積層膜であることも好まし い。このとき、高屈折率の層(第1層)が、金属酸化 物、金属硫化物または金属弗化物からなり、低屈折率の 層(第2層)が金属酸化物、金属弗化物または酸化ケイ 素からなる反射防止膜でであるのが好ましい。

【0058】さらに本発明では、反射防止膜が、酸化ケ イ素(SiOまたはSiO2)層を成形体と接する層(第1 層)とし、ついで高屈折率の層(第2層)および低屈折 率の層(第3層)を有する積層膜であると、特に成形体 との密着性が高く好ましい。このとき、高屈折率の層 (第2層)が、金属酸化物、金属硫化物または金属弗化 物からなり、低屈折率の層(第3層)が金属酸化物、金 属弗化物または酸化ケイ素からなる反射防止膜であるの が好ましい。

【0059】反射防止膜を形成することのできる、金属 酸化物、金属硫化物および金属弗化物としては、通常の 40 大気中もしくは水中で安定な物質であることが好まし い。金属酸化物の例としては、酸化アルミニウム、酸化 ジルコニウム、酸化セリウム、酸化ビスマス、酸化クロ ム、酸化鉄、酸化ハフニウム、酸化タリウム、5酸化タ ンタル、酸化イリジウム、酸化タングステン、酸化モリ ブデン、酸化ランタン、酸化マグネシウム、酸化サマリ ウム、酸化アンチモン、酸化ケイ素、酸化スカンジウ ム、酸化スズ、酸化チタン、酸化イットリウム、酸化ユ ーロピウム、酸化亜鉛などが挙げられる。

【0060】金属硫化物の例としては、硫化亜鉛、硫化 本発明の光学部品は、上述した環状炭化水素系重合体か 50 ロジウムなどを挙げることができる。金属弗化物の例と

しては、弗化アルミニウム、弗化バリウム。弗化セリウム、弗化カルシウム、弗化ランタン、弗化リチウム、弗化マグネシウム、クリオライト、チオライト、弗化ネオジウム、弗化ナトリウム、弗化鉛、弗化ストロンチウムなどが挙げられる。

【0061】反射防止膜の各層を形成するこれらの原料素材は、適宜混合して用いてもよい。このような反射防止膜は、特定波長の反射を防止する Vコート膜であっても、広帯域の反射を防止するマルチコート膜であってもよい。

【0062】このような反射防止膜を成形体上に形成する方法は、特に限定されるものではなく、膜の原料素材にもよるが、たとえば、真空蒸着法、反射蒸着法、イオンプレーティング法、スパッタリング法、イオンビームアシステッド法などが挙げられる。また、反射防止膜が複数の層からなる積層膜である場合には、これらの方法を適宜組み合わせて形成してもよい。また、本発明では、成形体にシリコン系樹脂によるアンダーコート処理を施した後、反射防止膜を形成してもよい。

【0063】本発明に係る光学部品は、反射防止膜の厚さを特に限定するものではないが、全膜厚で、通常10~500nm程度であるのが好ましい。本発明に係る光学部品は、目的とする波長の光を80%以上、好ましくは85%以上、特に好ましくは90%以上透過するのが望ましい。

#### [0065]

【発明の効果】本発明によれば、光学特性に優れ、かつ、耐久性に優れた反射防止膜を有する光学部品を提供することができる。このような光学部品は、光線透過率が高く、反射防止性能に優れ、かつ耐久性に優れた反射防止膜を有するため、種々の光学部品用途に好適に使用することができる。

#### [0066]

【実施例】以下、実施例に基づいて本発明をさらに具体的に説明するが、本発明はこれらの実施例に限定されるものではない。

#### [0067]

【調製例1】環状炭化水素系重合体(A)の調製 MFRが3g/10分(温度:200℃、荷重:5kg) であるポリスチレン(日本ポリスチ(株)製、G590)1 kgを、50リットルのシクロヘキサンに溶解し、さら に安定化ニッケル触媒N163A(日本化学工業社製、40 %ニッケル担持シリカ・アルミナ担体)150gを添加 50 して混合した。これを脱気処理した後、窒素ガスで置換し、更に 230  $\mathbb{C}$ 、50 k g/c  $m^2$  の水素圧で 10 時間水素添加反応を行った。

【0068】反応終了後、触媒を濾過により除去し、イソプロパノールで重合体を析出させた。このポリマーを濾過により分離し、減圧乾燥によりポリスチレンの水素添加物である環状炭化水素系重合体(A)を得た。

【0069】得られた水素添加物の物性は、Tgが143℃、密度が0.95g/cm³、水素添加率は99%以上であった。またX線回折による結晶化度は0%であった。

#### [0070]

【実施例1】調製例1で得た環状炭化水素系重合体 (A)を用い、シリンダー温度320℃の条件で射出成 形を行って、厚さ3mmの角板を得た。

【0071】この角板に、10-5 Torrの減圧条件下で、TiO2を65nm厚さに真空蒸着して反射防止膜を形成し、光学部品試料を得た。得られた光学部品試料について、反射防止膜の密着性および650nmにおける光線透過率を評価した。結果を表1に示す。

【0072】なお、反射防止膜の密着性は、反射防止膜を形成した光学部品試料を、80℃、相対湿度90%において48時間(条件1)・および、90℃、相対湿度90%において96時間(条件2)の各条件で静置した後、それぞれ粘着テープにより剥離テストを行い、目視により評価した。

#### [0073]

【実施例2】実施例1において、TiO2の代わりにCeO2を用いたことの他は、実施例1と同様にして光学部品試料を製造した。

【0074】得られた光学部品試料について、反射防止膜の密着性および650nmにおける光線透過率を実施例1と同様に評価した。結果を表1に示す。

#### [0075]

【実施例3】調製例1で得た環状炭化水素系重合体 (A)を用い、シリンダー温度320℃の条件で射出成 形を行って、厚さ3mmの角板を得た。

【0076】この角板に、10<sup>-5</sup> Torrの減圧条件下で、TiO2を65nm厚さに真空蒸着して第1層を形成し、次いでSiO2を4nm厚さに真空蒸着して第2層を形成することにより反射防止膜を形成し、光学部品試料を得た。

【0077】得られた光学部品試料について、反射防止膜の密着性および650nmにおける光線透過率を実施例1と同様に評価した。結果を表1に示す。

#### [0078]

【実施例4~9】実施例3において、TiO2を用いる代わりに、第1層として表1に示す化合物を用いたことの他は、実施例1と同様にして光学部品試料を製造した。

【0079】得られた光学部品試料について、反射防止

膜の密着性および650nmにおける光線透過率を実施

【比較例1】実施例3において、環状炭化水素系重合体

(三菱レーヨン社製、アクリペットVH) を用いたこと

膜の密着性および650nmにおける光線透過率を実施

【実施例10】調製例1で得た環状炭化水素系重合体

(A)を用い、シリンダー温度320℃の条件で射出成

【0083】この角板に、10-5 Torrの減圧条件下

で、SiO2を4nm厚さに真空蒸着して第1層を形成

し、次いでSiO2を65nm厚さに真空蒸着して第2

層を形成し、さらにSiO2を4nm厚さに真空蒸着し

て第3層を形成することにより反射防止膜を形成し、光

(A) を用いる代わりに、ポリメチルメタクリレート

の他は、実施例3と同様にして光学部品試料を製造し

例1と同様に評価した。結果を表1に示す。

例1と同様に評価した。結果を表1に示す。

形を行って、厚さ3mmの角板を得た。

[0080]

[0082]

学部品試料を得た。

た。

【0084】得られた光学部品試料について、反射防止 膜の密着性および650nmにおける光線透過率を実施 例1と同様に評価した。結果を表1に示す。

#### [0085]

【実施例11~15】実施例10において、TiO2を 用いる代わりに、第2層として表1に示す化合物を用い たことの他は、実施例10と同様にして光学部品試料を 製造した。

【0086】得られた光学部品試料について、反射防止 膜の密着性および650nmにおける光線透過率を実施 【0081】得られた光学部品試料について、反射防止 10 例1と同様に評価した。結果を表1に示す。

#### [0087]

【比較例2】実施例10において、調製例1で得た環状 炭化水素系重合体(A)を用いる代わりに、ポリメチル メタクリレート(三菱レーヨン社製、アクリペットV H)を用いたことの他は、実施例10と同様にして光学 部品試料を製造した。

【0088】得られた光学部品試料について、反射防止 膜の密着性および650nmにおける光線透過率を実施 20 例1と同様に評価した。結果を表1に示す。

#### [0089]

#### 【表 1】

					132 1 3		•
	成形体	反射防止膜			剥離テスト		光線透過率
	樹脂	第一層	第二層	第三層	条件 1	条件2	(%)
実施例1	(A)	TiO <sub>z</sub>	_	_	異常無し	一部剥離	95 以上
実施例2	(A)	CeO,		_	異常無し	一部剥離	95 以上
実施例3	. <b>(A)</b>	TiO <sub>2</sub>	SiO <sub>2</sub>		異常無し	一部剥離	95 以上
実施例4	(A)	CeO <sub>2</sub>	SiO,		異常無し	一部刺靡	95 以上
実施例 5	(A)	SnO <sub>2</sub>	SiO <sub>2</sub>	-	異常無し	一部剥離	95 以上
実施例6	(A)	ZrO,	SiO <sub>2</sub>		異常無し	一部剥艇	95 以上
実施例?	(A)	Ta <sub>2</sub> O <sub>5</sub>	SiO <sub>2</sub>	_	異常無し	一部剥離	95 以上
実施例8	( <u>A</u> )	WO <sub>3</sub>	SiO <sub>2</sub>		異常無し	一部剥離	95 以上
実施例 9	(A)	ZnS	SiO <sub>2</sub>	_	異常無し	一部剥離	95 以上
比較例1	PMMA	TiO <sub>2</sub>	SiO <sub>2</sub>		一部剥離	剥離	95 以上
<b>実施例 10</b>	(A)	SiO <sub>3</sub>	TiO,	SiO <sub>2</sub>	異常無し	異常無し	95 以上
実施例 11	(A)	SiO <sub>2</sub>	CeO,	SiO <sub>2</sub>	異常無し	異常無し	95 以上
<b>実施例 12</b>	(A)	SiO <sub>2</sub>	SnO <sub>2</sub>	SiO,	異常無し	異常無し	95 以上
実施例 13	(A)	SiO <sub>2</sub>	ZrO,	SiO <sub>2</sub>	異常無し	異常無し	95 以上
<b>实施例 14</b>	(A)	SiO <sub>2</sub>	Ta <sub>2</sub> O <sub>5</sub>	SiO <sub>2</sub>	異常無し	異常無し	95 以上
実施例 15	(A)	SiO <sub>2</sub>	WO <sub>3</sub>	SiO,	異常無し	異常無し	95 以上
比較例2	PMMA	SiO <sub>2</sub>	TiO,	SiO <sub>2</sub>	一部剥離	刺離	95 以上

:環状炭化水素系重合体(A)

PMMA : ポリメチルメタクリレート (三菱レーヨン社製、アクリペットVH)

光線透過率 測定波長: 650nm

剥離テスト 条件 1: 80°C 90% ×48hr 後 テープ剥離試験 剥離テスト 条件2: 90°C 90% ×96hr 後 テープ剥離試験

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